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RFP-INV-10
July 9, 1971

COMMITTEE EVALUATION OF PLUTONIUM LEVELS IN SOIL WITHIN AND SURROUNDING USAEC INSTALLATION AT ROCKY FLATS, COLORADO

J R. Seed, Chairman

K. W. Calkins

C. T. Illsley

F. J. Miner

J B Owen

"REVIEWED FOR CLASSIFICATION
By R B Hoffman
Date 7-11-97

REVIEWED FOR CLASSIFICATION/UCNI
By George H Sutherland
Date 10/14/08

THE DOW CHEMICAL COMPANY
ROCKY FLATS DIVISION
P. O BOX 888
GOLDEN, COLORADO 80401

U S ATOMIC ENERGY COMMISSION
CONTRACT AT(29-1)-1106

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Prepared under Contract AT(29-1)-1106
for the
Albuquerque Operations Office
U S Atomic Energy Commission

[redacted]

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ABSTRACT

On August 19, 1970 a committee was appointed by Lloyd M Joshel General Manager of the Rocky Flats Division of Dow Chemical U S A , to assess the long term potential hazard of plutonium contaminated soil under and around an asphalt pad at Rocky Flats The committee was also to make recommendations for disposition of the contaminated soil The area covered by the asphalt pad had been used as an outside storage area for plutonium contaminated oil drums This report contains the essential information relating to the evaluation The evaluation indicated that there is no health hazard associated with the levels of plutonium soil contamination found in the vicinity of the asphalt pad The report reviews the pertinent published literature and presents the results of research initiated in support of the investigation

[redacted]

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PART I STATUS

1 INTRODUCTION

In July 1958 at the USAEC Rocky Flats Installation an area on the plant site was designated as a temporary storage area for contaminated oil drums Subsequently some of the drums developed oil leaks and some plutonium contaminated oil was deposited on the soil The area was later covered by an asphalt pad

After a fire on May 11 1969 at Rocky Flats studies were conducted by the Health and Safety Laboratory (HASL) of the USAEC and by the Colorado Committee on Environmental Information (CCEI) concerning the possible release of plutonium These investigations detected measurable amounts of plutonium in the soil around the Rocky Flats Plant The epicenter quite clearly shows that this contamination could not be attributed to the May 1969 fire but is due to the resuspension and redistribution of contaminated soil from the oil drum storage area

A Committee was appointed by Lloyd M Joshel General Manager of the Rocky Flats Division of Dow Chemical U S A on August 19 1970 to assess the long term potential hazard and make recommendations for disposition of the plutonium contaminated soil under and around the asphalt pad, which covered the area that had served as an outdoor plutonium contaminated oil drum storage area

Members of the committee assembled over 3 000 pages of pertinent published literature Contacts were established with personnel throughout the AEC complex having similar problems and responsibilities (plutonium in soil)

In general the committee assembled and studied reports concerning

1 Rocky Flats historical information on the oil drum storage area

2 The HASL Report on *Plutonium in Soil Around the Rocky Flats Plant*, August 1 1970

3 Colorado State Department of Health soil survey data

4 A CCEI report by E A Martell entitled Report on the Dow Rocky Flats Fire Implications of Plutonium Releases to the Public Health and Safety January 13 1970

5 The Palomares Accident

6 The Thule Accident

7 Operation Plumbbob

8 Operation Roller Coaster

9 Soil analytical techniques

10 Techniques for plutonium removal from soil and contaminated soil disposition

11 Plutonium biological assimilation data

12 U S and other governments' recommended limits for plutonium contamination

13 Techniques for soil stabilization

14 Radiation survey instruments whose sensitivity and discrimination allowed for detection of plutonium-surface contamination

[redacted]

15 Reports on world wide fall out

Specific programs were initiated to evaluate the plutonium contamination data available from HASL and CCEI reports as well as the soil analysis data being collected by the Rocky Flats Health Physics Department Action was initiated to core sample and analyze the soil covered by the asphalt pad Action was also initiated to evaluate soil stabilization techniques Contacts were established with some of the other AEC sites that had potential problems with plutonium contamination in soil

The committee directed special attention to contamination standards that had been used in previous incidents of plutonium contamination in soil particularly the most recently proposed standards These standards were used in assessing the potential consequences of the level of plutonium contamination originating from the Rocky Flats oil drum storage area

2 ASSESSMENT OF POTENTIAL HAZARD

A reasonably thorough reading of the literature available on biological assimilation redistribution deposition and effects does not reveal any health hazard associated with the levels of plutonium soil contamination found in the vicinity of the asphalt pad There are unanswered questions relative to high LET radiation associated with low level internal exposure This is an area of radiation research which is being actively pursued by Los Alamos Scientific Laboratory, Battelle Northwest Lovelace Foundation Lawrence Radiation Laboratory the University of Utah, and the University of Rochester

The standards that have been used for plutonium soil contamination are based on sets of assumptions including resuspension data and an acceptable risk of 1 5 REM/yr to the adult pulmonary lumph nodes Although specific standards have been recommended in the past disaster or emergency situations have not used specific guidelines but rather a process of judicious decision making ' has been employed The main guideline for the "judicious decision making has been to maintain an "actual dose as near zero as possible ' This ' freedom" of interpretation, however, is shortly coming to an end It is this committee's opinion that the last data in the following table have been reasonably developed are realistic, and have a high probability of being adopted by the USAEC for plutonium in soil The assessment of hazard and our recommended actions should be consistent with these interim recommended standards It should be noted that the "contour lines ' established using the HASL Report 235 data and the data from the CCEI report, as well as the data

MAXIMUM PERMISSIBLE ALPHA CONTAMINATION

Country	$\mu Ci/m^2$	mCi/km^2 [a]	d/m/gram [b]	Remarks
United Kingdom (Dunster)	0 1	100	22 2	Widespread areas contaiminated with plutonium
United Kingdom	0 1	100	22 2	"Inactive areas "
	1 0	1000	222	"Active areas '
Czechoslovakia	0 11	110	24 4	Workplaces after decontamination
France	0 1	100	22 2	Equipment and workplaces in "inactive" areas
	1 0	1000	222	Equipment and workplaces in 'active" areas
Poland	0 1	100	22 2	Labs restricted to using 100 μ Ci or less
	1 0	1000	222	Labs permitted to use more than 100 μ Ci
South Africa	0 1	100	22 2	Body, personal clothing, inactive areas
	1 0	1000	222	Equipment and workplaces inside controlled areas
United States ICC	0 02	20	4 44	Interstate Commerce Commission (Dept of Transportation) pertains to interior of vehicles previously used for transportation of materials
U S S R	0 015	15	3 33	Work clothing and surfaces before cleaning
	0 002	2	0 444	Hands and work underclothing before cleaning
	0 006	6	1 33	Work Surfaces after cleaning
United States [c]	0 04	40	8 8	Urban suburban, recreation areas
	0 4	400	88	Rural truck farming, annual food crops, grazing land milk-shed, etc
	4 0	4000	888	Rural deep root perennials (e g nuts, certain fruits)
	40 0	40000	8888	Remote or Controlled desert, forest, fenced or limited access areas

[a] Units used in HASL report No 235
[b] Units used by Rocky Flats in reporting soil analysis (in most cases a specific gravity of one (1) was assumed for conversion of units
[c] Recommended at an International Symposium on Radiological Protection of the Public in a Nuclear Mass Disaster (June 1968)

obtained by the Rocky Flats Health Physics Department show values in excess of the recommended limits extending beyond our property boundaries

Other significant regulations that should be considered in assessment of hazard are

1 Criterion I of 10 CFR 140 84 (Financial Protection Requirements and Indemnity Agreements) which considers surface contamination values above 0 35 $\mu Ci/m^2$ (77 dpm/cm^2, 350 mCi/km^2) over 100 m^2 of property as the minimum level of transuranic radio nuclides that would comprise a substantial discharge of radioactive material from its intended place of confinement In the HASL report there are contour lines extending beyond the plant boundary that show plutonium concentrations in excess of this value

A Colorado State Board of Health Regulation should be considered

Radiation Regulation No 2

Requiring stabilization of uranium and thorium mill-tailing piles This regulation applies to *all* uranium and thorium mill tailing piles and could be interpreted by the courts to apply to our radioactive "pile It states that all such piles "shall be stabilized against wind and water erosion It states that any recognized technique might be employed such as concrete petroleum products etc so as to ensure proper protection from wind and water erosion Access to the stabilized area shall be controlled by the operator or owner "

Although the plutonium contamination of soil in and near Rocky Flats can not be considered a "mill tailing pile," this Regulation gives an indication of the State's thinking in a somewhat analogous situation

Initial estimates made by Rocky Flats Manufacturing personnel based on total number of leaking barrels and average plutonium concentration in the oil led to a value of approximately 85 grams total plutonium deposited in the soil beneath the barrels

Making use of all the soil analysis data, one can calculate an estimate of 7 6 grams over 5 63 km^2 (1,393 acres) outside the plant boundary down to the 13 mCi/km^2 contour The authors of the HASL 235 report estimate between 2 6 and 5 8 Ci off AEC property (38 to 85 grams Rocky Flats plutonium)

After taking all aspects into consideration the committee concludes that additional land should be purchased as proposed separately

3 HISTORY OF PLUTONIUM CONTAMINATED OIL DRUM STORAGE AREA

From the beginning of operations of the Rocky Flats Plant organic liquids contaminated with radioactive materials were generated in various manufacturing operations In the initial design of the facilities very little attention was given to this particular radioactive waste problem The volumes were very low and it had been assumed that this form of contaminated waste could be either burned or packaged in some manner and shipped for burial as were the low level solid wastes In the early days of the Rocky Flats operation some uranium contaminated oil was buried and some incinerated In 1957 a small quantity of uranium-contaminated oil was shipped to the Idaho burial site but shipment of contaminated liquid wastes was not continued

Changes in weapons design and in manufacturing processes significantly increased the amount of contaminated oils being generated This particularly significant in the manufacturing of plutonium components The problems of permanent disposals and of storage of the increasing quantities generated were recognized in 1956

As a result of one study the Part IV addition (completed in 1957) to the plant included a high speed centrifuge in Building 776 to process plutonium contaminated organic liquids The operation was disappointing and resulted in a recommendation made in 1958 that a substitute process be developed for disposal This investigation was initiated immediately by the Technical Staff predecessor of the existing Research and Development Organization at Rocky Flats Considerably more comprehensive than the earlier studies the processes investigated for disposal included distillation steam stripping ion exchange filtration ion exclusion evaporation solvent extraction and other approaches

The outside plutonium contaminated oil drum storage area was first established in July 1958

Most of the drums transferred to the field were nominal 55-gallon drums but a significant number were 30-gallon drums Not all were completely full Approximately three-fourths of the drums were plutonium-contaminated whereas most of the balance contained uranium Of those containing plutonium most included lathe coolant consisting of a straight-chain hydrocarbon mineral oil (Shell Vitrea) and carbon tetrachloride in varying proportions Other liquids were involved however including hydraulic oils vacuum pump oil trichloroethylene perchloroethylene silicone oils acetone still bottoms etc Originally contents of the drums were indicated on the outside but some of these markings became illegible through weathering and adequate records were not kept of the specific contents of each barrel Leakage of the oil was recognized early and in 1959 ethanolamine was added to the oil to reduce the corrosion rate of the steel drums

Development work on a potential process to dispose and/or reclaim the materials continued As a result of the development studies which had been initiated however a recommendation was issued in December of 1959 that a still be constructed for the separation purification, and reuse of the carbon tetrachloride and the Shell Vitrea A process design was forwarded to Plant Engineering The process was set up in Building 771 Because of time and funding problems surplus stainless steel equipment was used On May 15 1960 test runs on this equipment were begun and shortly afterward drums of currently generated oil together with some transferred from the field were processed through the system

Concurrently processes to dispose of the still bottoms from this operation and of other liquids were being pursued with incineration receiving the most favorable attention In this system the waste heat would be utilized to evaporate aqueous wastes which were also beginning to be a problem

In June of 1960 corrosion of the stainless equipment caused by hydrolysis of the CCl_4 to HCl became a problem and in September the operation was discontinued because of severe corrosion

After additional development work to solve the corrosion and other problems a revised design was submitted to Engineering in December 1961 The process was included in the project Additional Processing Facilities Contract AT(29-2)-1298' which was an expansion of the plutonium chemical operation During this period development of a sludging process for disposing of the still bottoms and other wastes by mixing with an activated silica was also completed In June 1963 a decision was made to delete the CCl_4 still and other features from the expansion project because of funding problems Because of this, the design capacity of the sludging process was increased to provide for processing all contaminated liquids and funds for this project-based on a mixer-extruder system called the "jelly factory' were requested Installation of the mixer-extruder system was completed in January 1964, but start-up work revealed major deficiencies which required extensive modification in the installation These modifications were not completed until late in 1965

When this equipment was finally ready for operation a further delay was encountered when it was determined desirable to provide a facility to perform the dual purposes of transferring the oil to new drums for site transfer to the mixing equipment and filtering through one micron filters for plutonium removal and recovery. As a result FY-1966 expense funds were provided to build a temporary facility for these purposes.

After more start up problems, the final phase of emptying the drum field began on January 23, 1967. By this time the field contained about 5,240 drums, of which approximately 3,570 contained plutonium contamination. The oldest drums and those containing plutonium were processed first. To the best of our knowledge, the last of the plutonium contaminated oil was removed on January 25, 1968. The last of the uranium contaminated oil was transferred to a new drum on May 28, 1968, and shipped to the disposal plant in June 5, 1968.

Original estimates of plutonium content had indicated that the plutonium bearing drums averaged about 4.5 grams of plutonium per drum. The material balance after processing however showed that less than half this amount was present. Of the plutonium found, only 594 grams were recovered. 2471 grams were processed with the oil and 5152 grams remained in the emptied drums.

An estimate of leakage based upon a material balance around the drums indicated that 5,000 gallons of oil containing about 86 grams of plutonium leaked from the drums into the soil.

The significant or pertinent events associated with the Plutonium Contaminated Drum Storage Area can be summarized as follows:

Date		Information
July	1958	Drum storage area established. During subsequent years, drums were continually added which primarily contained plutonium contaminated machining oils.
July	1959	First drum leakage discovered. Rust inhibitor ethanolamine was added to drums prior to storage to minimize corrosion.
January	1964	First evidence of large scale deterioration of drums reported. Soil contamination reported as increasing.
January	1966	Small building added to filter and transfer contaminated oil from leaking drums to new drums.
January	1967	Last drums added to storage area and removal to 774 began. Oldest drums shipped first.
June	1968	Last drum shipped to Building 774 for processing. High winds spread some contamination.
July	1968	Radiation monitoring and mapping of area completed. Levels of 2×10^5 d/m/g to over 3×10^7 d/m/g reported. Penetration of from 1 inch to 8 inches reported.
September	1968	Preliminary proposal for containment cover prepared by Rocky Flats Facilities Engineering.
July	1969	First coat of fill material applied.
August	1969	Fill work completed, paving contract let.
September	1969	Overlay material, soil sterilant and asphalt prime coat completed.
November	1969	Asphalt containment cover completed including four sampling wells.

The deposition of the contamination in the soil of the drum storage area began shortly after the drums were placed in the area. Resuspension and redistribution of the contamination however was certainly not a simple mathematical function of time. The quantity redistributed was directly associated with removal of the drums which exposed the contaminated soil, physical activity in the area, and the periodic high winds found at Rocky Flats. This can best be seen if we study data from air samples collected directly east of the storage area. The Rocky Flats Health Physics Department conducts an extensive air sampling program to determine the presence of airborne radioactive contaminants in and around the Rocky Flats Plant site. One of these air samplers (designated S-8) is situated directly east of the 903 drum storage area at the east perimeter fence. The contamination levels determined by analysis of samples collected daily from this location have, through the years, been higher than the levels measured at other locations.

Monthly averages of the individual daily contamination levels show a very close correlation to physical activity in the drum storage area. Figure 3-1 shows a graph of the monthly average airborne contamination levels determined at air

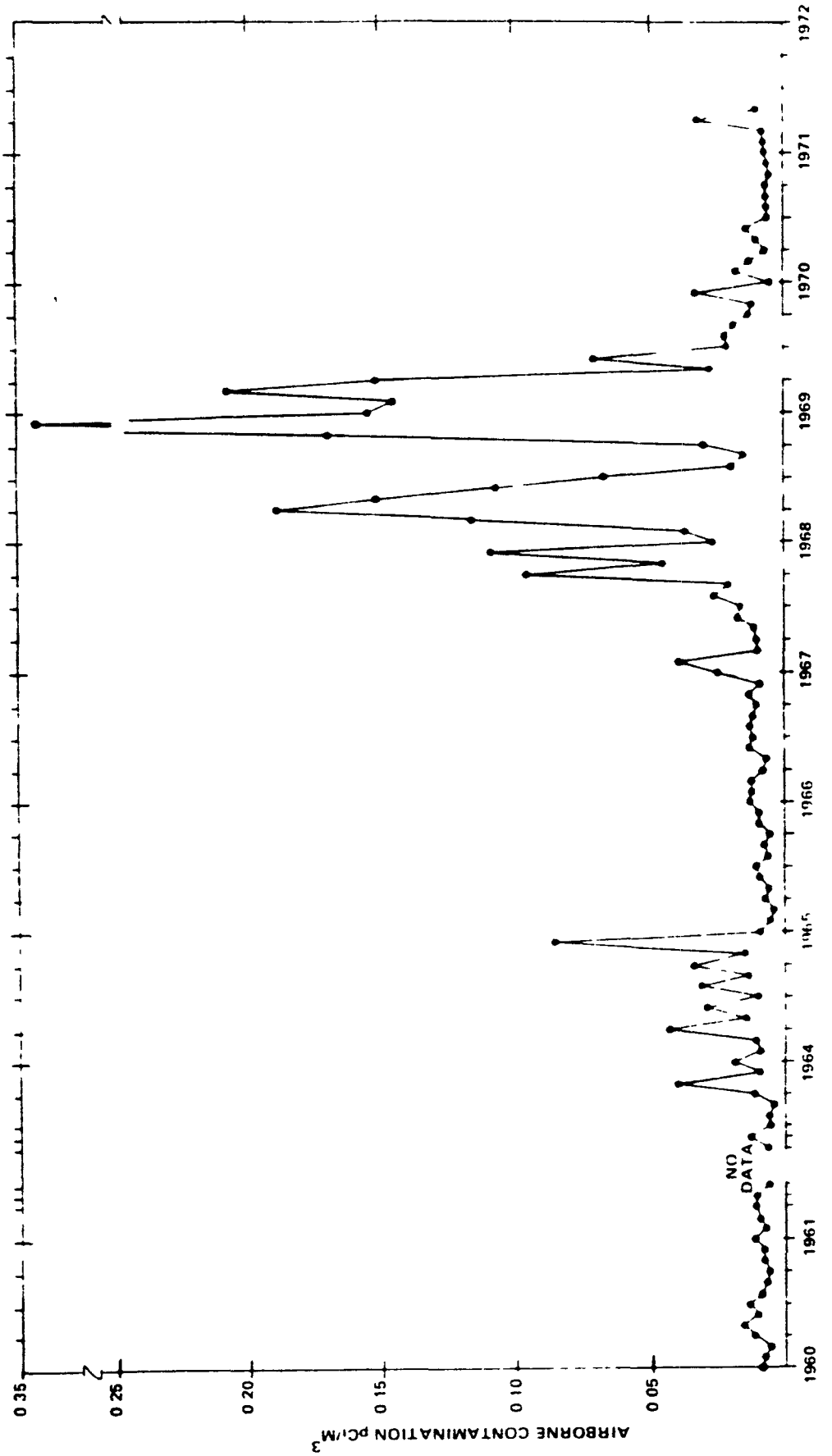


Figure 3 1 Monthly Averages of Daily Airborne Contamination Values from Air Sampler S 8 Located Just East of the Drum Storage Area

sample [illegible]. When analyzing these data it is worth noting that from [illegible] 1963 to 1965 large scale deterioration of the drums was taking place. An exclusion fence was added (to limit [illegible] physical activity in the area). One drum was punctured by a fork lift and some soil was added to cover certain contaminated areas. These physical activities corresponded to the higher readings reported during the 1963 through 1965 period. Starting in January 1967 a great deal of activity in the area was associated with drum removal. The contamination level measured in 1968 paralleled the drum removal activity. In late 1968 activity associated with weeds being burned and grading of the area can be correlated with high contamination readings. In April of 1969 when the large rocks and stakes were removed for filling and leveling the area increased contamination levels were reported.

In November of 1969 the four sample wells were dug and the increases in contamination level are attributed to the activity in the area. In April of 1971 a new drainage ditch was completed just west of this area and, again, higher contamination values were observed.

At no time did the values from air samples indicate that permissible levels of plutonium contamination in air had been exceeded. However these data do indicate the most probable period when maximum resuspension and redistribution of the contaminated soil took place.

4 ATOMIC ENERGY COMMISSION CONTACTS

To collect as much information as possible concerning contaminated soil problems knowledgeable individuals throughout the AEC complex were contacted The following is a summary of these contacts

LASL

Wright Langham (Health Division) discussed availability of reports on plutonium contamination of soils especially reports related to the Palomares and Thule accidents Langham reported that this information is not readily available because it had not yet been published by the Air Force Langham reviewed HASL report 235 for the AEC and said he was reasonably impressed that a professional job had been done He mentioned by telephone communications and later suggested in writing that the Dow committee might profit by a visit to Los Alamos to talk to people familiar with foreign contamination incidents as well as with contamination incidents at Los Alamos It was suggested that Dean Meyer John Healy and William Kennedy could provide information concerning contamination levels in the LASL area

USAEC

Philip Krey (HASL) discussed HASL report 235 Two errors one on the contour map of Rocky Flats area soil survey and one typographical error in the data table on plutonium concentration in soil at one sample site were corrected It was agreed that neither correction significantly changed the total plutonium inventory in the area surrounding Rocky Flats

John S Harley (HASL) mentioned that he had participated in a meeting at the AEC Headquarters on July 6 7 1970 to draw up guidelines for sampling and analyzing soils for plutonium Harley also discussed the possibility of HASL personnel returning to Colorado for a supplemental program to include (1) more detailed surveys in the immediate vicinity of Rocky Flats and (2) specific field studied on resuspension of plutonium contaminated soil particles

Don Ross (Health Protection – Operational Safety) discussed in great detail the AEC guidelines (prepared July 6 and 7) for plutonium sample collection and analysis This discussion with Ross reviewed the initial request from ALO (W B Johnston) for such a meeting the membership of the committee which developed the guidelines, how the guidelines were arrived at, and a summary of the nine guidelines This document has been routed to Dow through ALO

Art Whitman (NVO – Rad Op) discussed the availability of NVO reports It was suggested that we obtain a copy of a special bibliography on plutonium soil contamination (this bibliography has been received) Also Whitman suggested that someone from Dow visit NVO to discuss details of NTS plutonium-contamination experience and to scan their files of classified reports and miscellaneous data for information pertinent to the Rocky Flats situation Therefore a trip was taken to Nevada The following is a list of persons contacted at the Nevada Operations Office United States Public Health Service and Reynolds Electrical and Engineering Co A trip report summarizing this activity is given in Appendix E

USAEC

Arthur J Whitman	Radiological Safety Branch
Donald W Hendricks	Radiological Safety Branch
Ross L Kinnaman	Effects Evaluation Office
Paul J Mudra	Operations Division

USPHS - SWRHL

Mel W Carter	Laboratory Director
Jim Mullins	Deputy Chief Analytical Division
Les Dunn	Environmental Survey

REECO (Test Site)

Arden E Bicker	Environmental Services
Derek Engstrom	Chemistry Laboratory
Leonard Svgitowicz	Chemistry Laboratory
Terry Roy	Site Survey

ORNL

F R Bruce (Safety and Radiation Control) and W S Snyder (Health Physics) were contacted Discussions were held concerning a contamination release which ocurred in 1957 An explosion in a process tank allowed the release of a uranium-plutonium solution Most of the contamination was contained within a building However some contamination to the environment resulted by personnel tracking it outside and the escape of airborne contamination It was estimated that about 1 gram of plutonium was lost Some gravel and top soil were removed and placed in drums for disposal

NEVADA

C A Pelletier (Environmental Health Services Laboratory) stated that work was being conducted concerning the deposition of radioactive material from the air to soil However, resuspension of plutonium from the soil, and soil stabilization have not been investigated

Claude Sill discussed the Idaho method (fusion and total dissolution) for analysis of plutonium in the soil

5 REVIEW OF CURRENT ANALYTICAL METHODS FOR PLUTONIUM IN SOIL

Measuring plutonium in soils especially at very low levels is a difficult analytical problem It is imperative that the analytical methods have good accuracy precision and reproducibility of information

Various methods have been used for the analysis of plutonium in soils They differ primarily in the method by which the plutonium is released from the soil These methods vary from acid leaching to complete dissolution of the soil by fusion

The specific methods used by various sites for plutonium analysis have been collected Three of these (Rocky Flats Health Physics Laboratory and Building 881 Analytical Laboratory and the AEC's Health and Safety Laboratory) are summarized below Following these summaries there is a discussion of the reproducibilities of the various methods comments on the two Rocky Flats Laboratories that have the capabilities for analysis of plutonium in soils and a summary of the AEC guidelines for soil sampling and analysis

Methods Summary

1 Rocky Flats (Health Physics)

The soil samples (10g) are ignited a ^{236}Pu spike (used to measure the recovery of all the plutonium in the sample) is added and the samples are evaporated (leached) alternately with HNO_3 and HCl This is followed by a leach with HF and $HClO_4$ The solids are then removed by filtration the acidity of the solution is adjusted with HCl and the Pu is separated by anion exchange The plutonium concentration is then determined by alpha spectrometry

2 Rocky Flats (Building 881 Analytical Laboratory)

A ^{236}Pu spike is added to 100g of soil and the sample is leached twice with a 1 to 3 mixture of HCl and HNO_3 Plutonium is removed from the leaching solution on a column of anion exchange resin After stripping the plutonium from the resin with a dilute HNO_3 HF mixture the acidity and plutonium valence are adjusted and the plutonium is extracted with TTA The plutonium-containing organic solution is then evaporated on a planchet and the plutonium determined by alpha spectroscopy

3 AEC Health and Safety Laboratory (HASL)

A ^{236}Pu spike is added and the samples (100g) are leached with a HNO_3 HCl mixture first at room temperature then at an elevated temperature The solution is decanted and any organic matter is decomposed by heating Silicic material if present is removed by filtration and decomposed by heating with HF The remaining material is dissolved in HNO_3 and combined with the initial filtrate The acidity is adjusted with HNO_3 and the plutonium is separated using anion exchange The plutonium is then plated on a disk and the concentration determined by alpha spectrometry

Other methods for determining plutonium in soils involve complete dissolution of the soil by fusion, then separation of the plutonium from the fused mass In the AEC complex, Claude Sill of the Idaho Operations Office is a proponent of this method The primary drawback of the fusion method is the extensive time required to process each sample and the relatively small sample size (1 to 5g usually) that can be handled conveniently Those favoring the fusion method are convinced however, that it is the only sure way of getting all of the plutonium in solution and available for analysis

The amount of plutonium that will dissolve by leaching depends on its form in the soil In analytical methods ^{236}Pu "spikes are added in solution form As a result this 'spike" of plutonium will be more easily removed from soil than plutonium in the form of oxide especially oxide that is high fired Although there is probably no better way to spike a soil sample those using soil analysis data should be aware of the limitations and the data should be evaluated accordingly

Fuming a soil sample with an HF $HClO_4$ mixture is another method that has been used for complete soil dissolution It too is a lengthy process The use of this method has been reported by HASL (HASL 235) The current Rocky Flats method uses a HF $HClO_4$ acid treatment, but not to the point of complete dissolution of the sample

Reproducibility of Analytical Methods

There are no analytical standards for plutonium in soil The only way that analytical methods can be compared, therefore, is by percent recovery of spikes, the precision of these spike recoveries, and interlaboratory analysis of the same sample Data on spike recovery and the precision of this recovery are summarized below for the Rocky Flats and the HASL methods This is followed by a discussion of results on interlaboratory sample exchange programs and a comparison of different methods on the same samples

SPIKE RECOVERY

1 Rocky Flats Health Physics Laboratory

In mid 1970 a series of soils from LRL was analyzed at Rocky Flats The plutonium content ranged from 0 3 to 500 d/m/g with 5 samples ≤1 d/m/g one sample at 10 d/m/g and one sample at 500 d/m/g The average percent recovery of the spikes for all replicates was 76% with a standard deviation of 7%

2 AEC Health and Safety Laboratory (HASL)

J H Harley provided data on spike recovery for their method On ten soil samples analyzed the average recovery was 84% with a standard deviation of 6% The data supplied were in the form of spike recoveries only the amount of plutonium in the samples was not reported

Comparison of Different Analytical Methods

HASL has compared their leach method a carbonate fusion method and a HF $HClO_4$ dissolution method all on Rocky Flats soils The results are shown in Tables 5 1 and 5 2 These results indicate that the methods are reasonably comparable (not significantly different according to the HASL report) It should be noted however that there is no indication of the chemical form of plutonium in the soil used to obtain the results in either of these tables

Table 5-1 Comparison of Plutonium Analysis in Soil by Nitric Hydrochloric Acid Leach and Sodium Carbonate Fusion Methods

Soil Type	Method	dpm/100g Pu 239 + 240	dpm/100g Pu 238	Percent Chemical Yield
1A	Leach	308 ±11	5 9 ±0 2	53
1B	Fusion	318 ±12	6 6 ±0 3	59
2A	Leach	1629 ±87	32 ±2	56
2B	Fusion	1607 ±82	35 ±2	66
3A	Leach	6 0 ±0 2	0 19 ±0 01	58
3B	Fusion	8 0 ±0 3	0 13 ±0 01	61

This information received from J H Harley HASL September 3 1970

Table 5 2 Complete Dissolution versus Leaching

Map Site	Sample	Rocky Flats Soils Lab	Method	Pu 239 dpm/g ±2 σ	% Deviation Between Methods
7	14	HASL	leach	3 08 ±4	3 2
		HASL	fusion	3 18 ±4	
6	6	HASL	leach	18 7 ±4	
		HASL	leach	16 3 ±5	8 9
		HASL	fusion	16 0 ±5	
4	4	HASL	leach	0 060 ±4	28
		HASL	fusion	0 080 ±4	
				mean 13	
		Other Soils			
N Y C 6/67	0 20 cm	IPA	leach	0 0167 ±4	
		IPA	leach	0 0165 ±5	0
		HASL	fusion	0 0166 ±4	
N Y C 12/69	0 2 5 cm	TLW	leach	0 22 ±10	
		TLW	leach	0 20 ±5	11
		TLW	HF dissolution	0 23 ±8	
		TLW	HF dissolution	0 24 ±5	
N Y C 12/69	0 5 cm	TLW	leach	0 091 ±6	
		TLW	leach	0 096 ±5	3 3
		TLW	HI dissolution	0 090 ±6	
		TLW	HI dissolution	0 092 ±5	
N Y C 12/69	5 20 cm	TLW	leach	0 0048 ±20	
		TLW	leach	0 0041 ±16	7 1
		TLW	HI dissolution	0 0035 ±42	
		TLW	HI dissolution	0 0049 ±13	
		HASL	fusion	0 0041 ±5	
Black Soil 1958		TLW	leach	0 0074 ±35	37
		HASL	fusion	0 0051 ±4	
				mean 12	

Comparison of Interlaboratory Results

LRL supplied analyses on the samples they sent to Rocky Flats and mentioned above The results are tabulated below Qualitatively the agreement between the two series of results is quite good

Sample Number	d/m/g LRL	d/m/g Rocky Flats[a]
1	0 06	0 29 ±0 19
2	0 67	0 82 ±0 22
3	0 4	0 71 ±0 22
4	0 2	0 44 ±0 15
5	0 14 0 91	1 08 ±1 07
6	550	493 ±33
7	13	10 9 ±0 8

(a) $\text{mean} \pm \frac{\text{range}}{2}$

The Rocky Flats Health Physics Laboratory was involved in an interlaboratory sample exchange in mid 1970 using samples prepared by Claude Sill of Idaho The soil samples were apparently fused after preparation so that the plutonium was present in the form of a high fired oxide On the first two of these there was a recovery of 65 and 100% (the analytical method used includes leaching with HF which will dissolve high-fired oxides better than any other treatment) This recovery was better than that obtained by either HASL or Los Alamos Subsequently two other Sill-prepared samples were analyzed The Laboratory obtained an 85% recovery on a sample containing 23 d/m/g and a 71% recovery on a sample containing 39 d/m/g

Currently the Rocky Flats Health Physics Laboratory is involved in a ^{239}Pu Soil Cross-Check Program being administered by the Southwestern Radiological Health Laboratory Five samples have been received and three were analyzed The concentration of plutonium in these samples is in the range of 5 to 30 d/m/g Verbal information has been obtained on the first sample analyzed and indicates a recovery of between 95-100% of the plutonium present (~33 d/m/g) Three aliquants were run The precision as measured from the three aliquant results was ±5%

HASL has conducted interlaboratory comparison of methods They are summarized in Table 5 3 For the most part the results from the various laboratories appear to be within reasonable agreement

Table 5 3 Interlaboratory Comparison Analysis of Plutonium in Soil

Sample Location	Lab	Method	dpm/100g Pu 239 + 240	dpm/100g Pu 238
Colorado (1)	A	HNO_3, HCl Leach	400 ±4	7 2 ±0 2
	HASL	HNO_3, HCl Leach	308 ±11	5 9 ±0 2
	HASL	Na_2CO_3 Fusion	318 ±12	6 6 ±0 3
Colorado (2)	A	HNO_3, HCl Leach	1660 ±33	31 8 ±1 3
	HASL	HNO_3, HCl Leach	1629 ±87	31 8 ±2 0
	HASL	Na_2CO_3 Fusion	1607 ±82	35 3 ±1 6
Colorado (3)	A	HNO_3, HCl Leach	10 2 ±0 2	0 20 ±0 03
	HASL	HNO_3, HCl Leach	6 0 ±0 2	0 20 ±0 01
	HASL	Na_2CO_3 Fusion	8 0 ±0 3	0 13 ±0 01
New York (1)	A	HNO_3, HCl Leach	1 7 ±0 1	0 04 ±0 01
	HASL	Na_2CO_3 Fusion	1 7 ±0 1	0 41 ±0 02
New York (2)	B	HNO_3, HCl Leach	0 41 ±0 07	0 27 ±0 21
	B	HF Dissolution	0 35 ±0 15	0 09 ±0 18
	B	HF Dissolution	0 49 ±0 07	0 04 ±0 04
	HASL	Na_2CO_3 Fusion	0 41 ±0 02	0 03 ±0 02
Illinois (1)	B	HNO_3, HCl Leach	0 74 ±0 25	–
	HASL	Na_2CO_3 Fusion	0 51 ±0 03	–

Table 5 3 (continued)

Sample Location	Lab	Method	dpm/50g Pu 239 + 240	dpm/50g Pu 238
New York (3)	B	HNO_3, HCl Leach	4 82 ±0 23	0 03 ±0 03
	B	HNO_3, HCl Leach	4 53 ±0 30	0 39 ±0 10
	B	HF Dissolution	4 58 ±0 25	0 20 ±0 10
	B	HF Dissolution	4 49 ±0 24	0 14 ±0 09
	HASL	HNO_3, HCl Leach	4 13 ±0 17	0 32 ±0 02

General Comments

ROCKY FLATS HEALTH PHYSICS LABORATORY

The laboratory personnel are currently investigating a total dissolution method using a Teflon-lined Parr Bomb Pressures as high as 1200 psi can be obtained in this bomb It is hoped that with this high pressure and a mixture of HF and HCl it will be possible to dissolve a greater fraction of the sample

The back log of samples in the Laboratory is being reduced The average elapsed time for a sample analysis is now one month however the time depends on the priority of the sample It takes about a week from the time analytical work begins on a sample until the results are available

BUILDING 881 ANALYTICAL LABORATORY

The Building 881 Analytical Laboratory has been developing a capability for plutonium analysis in soils over the last six to eight months They currently have a method developed [Methods Summary (2)] and have obtained the necessary counting equipment Analyses of soil samples taken from the Pad are in the range of 0 001 to 7 7 μg/100 g soil They estimate their precision on these samples to have been ±20%

To investigate the effect of the form of plutonium on its recovery they spiked a large soil sample with plutonium and heated it to 400° 600°, 800° and 1000°C They analyzed samples heated to each temperature by their method (which includes an HCl-HNO_3 leach) and recovered 105, 109 85, and 13%, respectively, of the plutonium This graphically demonstrates the influence that the form of plutonium has on its recovery This is an important observation The nature of the plutonium in the soil sample should dictate the dissolution technique used If is is suspected that a "high-fired" oxide is involved, the fusion method recommended by Claude Sill (or near complete dissolution) might be necessary for reasonable quantitative results

AEC Guidelines for Soil Sampling and Chemical Analyses

It is apparent from the information above that there is not general agreement between various sites on the best analytical method to use for soil analysis. The same situation exists for soil sampling techniques.

During the course of this committees investigation the AEC issued a set of guidelines for soil sampling and sample analysis (copy received October 26, 1970). Criteria are given for determining locations for sample taking, the number and frequency of samples to be taken, the procedure for taking the samples, and the preparation of the samples for subsequent analysis. Rocky Flats procedures (within the limits imposed by the nature of our rocky soil) have taken cognizance of these guidelines.

The criteria for sample analysis are general and the methods now in use appear to comply with the AEC recommendation.

6 SIGNIFICANT, PERTINENT INFORMATION FROM PREVIOUS INCIDENTS OF PLUTONIUM CONTAMINATION

There have been several instances of plutonium contamination of the environment outside of a government (AEC) controlled area. Two of these – Palomares, Spain, January 1966 and Thule, Greenland, January 1968 – involved major plutonium contamination of significant land areas. There were also significant political and public relation overtones. It is instructive to examine these two accidents to see what can be learned about the extent of contamination, the methods used to decontaminate, and the amount of residual activity remaining after decontamination was completed. The Palomares accident will be examined first, since it more closely resembles the Rocky Flats situation with contaminated soil.

Palomares

EXTENT OF CONTAMINATION

The accident at Palomares took place at 32 800 feet with pieces of the plane falling over a very wide area. Plutonium was released by the non-nuclear explosions of two bombs. A total of approximately 558 acres was contaminated. A total of 5.4 acres had an alpha contamination of more than 700 000 d/m/100 cm^2 (See Appendix D for simplified conversion to other common units found in this report.) An area of approximately 42 acres showed contamination of between 700 000 and 70 000 d/m/100 cm^2, and the rest, some 511 acres, showed contamination of less than 70 000 d/m/100 cm^2. More than half of these 511 acres were contaminated to less than 7 000 d/m/100 cm^2.

DISPOSITION OF CONTAMINATED SOIL

The disposition of the contaminated soil is summarized as follows:

DISTRIBUTION OF PLUTONIUM ON THE SURFACE IN THE VICINITY OF THE PALOMARES ACCIDENT

Initial Plutonium Contamination			Area	
$\mu Ci/100\ cm^2$	mCi/km^2	d/m/100 cm	acres	Disposition
>0.32	>32 000	>700 000	5.4	Surface soil (2.3") removed and buried at Savannah River
0.32-0.032	32 000 3 200	700 000 70 000	42 [a]	Deep plowed, watered, and some vegetation removed
<0.032	<3 200 Detectable	<70 000	511 [b] 558	Deep plowed and watered

[a] From one report it could be concluded that this soil was also removed to Savannah River for burial, but this does not seem likely when we analyze all the reports on this topic.

[b] Rather more than half is less than 0.0032 $\mu Ci/100\ cm^2$.

The removal of soil from the 5.4 acres where the activity was highest resulted in 1100 cubic yards of soil which were buried at Savannah River in the same manner as other low level radioactive material. (To help gain a perspective, the soil buried contained about 10^{-3} to 10^{-4} times the amount of plutonium found in the remaining area.) Also removed from the site and buried at Savannah River were about 400 cubic yards of vegetation. It was planned initially to deep plow only 300 acres of land having low but discernible amounts of contamination. However, the operation was found to be so easily performed that the area was extended to include an additional 558 acres. Plowing was done to a depth of approximately 10 to 15 inches, the soil was primarily sandy although some places were rocky. This process reduced the surface contamination in this area to undetectable amounts and it was concluded, essentially eliminated significant resuspension of plutonium into the air.

The total cost for the operation in Spain has been estimated as 50 million dollars. This does not include the political and public relations "cost" of the incident.

A comparison can be made between the quantities of plutonium found in the soil at Palomares and the amount found in the vicinity of the barrel storage area at Rocky Flats. This comparison should be considered only an order-of magnitude comparison since details of soil sampling at Palomares are not available to us. An assumption is made that the reported results from Palomares are from surface sampling only. At Palomares soil was actually removed (and shipped to the United States for burial) where contamination levels were greater than 0.32 $\mu Ci/100\ cm^2$ (32 000 mCi/km^2). All soil containing measurable contamination up to the value of 0.32 $\mu Ci/100\ cm^2$ was deep plowed. Data from the HASL Report 235 indicated that the two hottest spots found at Rocky Flats were east of the barrel storage area. These spots (indicated as sites 6 and 8 in HASL Report 235) contained totals of 2000 and 620 mCi/km^2, respectively, in the 0-20 cm depth. Of the total plutonium reported from these sampling sites, 1320 and 415 mCi/km^2 were reported as occurring in the 0-5 cm depth. Rocky Flats Health Physics Department sampling and analysis give results in excess of 3 000 mCi/km^2 on AEC land. The highest isocurie contour that extends on private property surrounding AEC land was found to be between 400 to 1000 mCi/km^2. The plutonium contamination levels of concern at Rocky Flats are substantially below those levels at Palomares where soil removal was deemed necessary.

CURRENT LEVELS OF CONTAMINATION

Sampling of the soil has been done on a yearly schedule in the Palomares region since the accident in 1966. Core samples are taken in areas that had varying initial contamination. These cores are divided into segments of 5 to 10 cm

for a total depth of 45 cm The conclusions reported at a conference in 1968 are as follows

1 In the area where the 2 3 inch layer of contaminated soil was removed and sent to Savannah River the contamination is nil

2 In the areas where there was deep plowing contamination has been found to a depth of 13 inches Generally highest contamination levels were found between 6 and 10 inches down but there was a very unhomogeneous distribution of the contamination

3 The maximum average value of alpha activity found in the areas studied is approximately 50 times higher than the minimum value of natural alpha activity found in the background soil in the area

A network of four air-sampling stations was set up within the accident area Air samplers were placed 5 5 feet above the ground They operated 24 hours a day throughout the year Cellulose filters with a pore size of 1 2 microns were used for the sampling Gross alpha as well as ^{239}Pu content were determined on the filters The results of these air samples were reported at meetings in 1968 and 1970 They are summarized as follows

1 Activity has been found at all sampling areas This means that some radioactive material has become resuspended However removal of the most contaminated surface soil and the dilution of the remainder by plowing has proven effective in reducing the mean value for ^{239}Pu in the air to levels consistently below the permissible maximum

2 The ^{239}Pu concentration in the air was normally less than 0 1 of the maximum permissible concentration (MPC) for the general public (The MPC used was from the "Radiation Protection Norms of the European Nuclear Energy Agency revised edition 1968 For insoluble compounds of ^{239}Pu in air for the general public, it is 10^{-12} $\mu Ci/cm^3$ for soluble compounds it is 6×10^{-4} $\mu Ci/cm^3$ Although the plutonium compounds are known to be insoluble plutonium dioxide the soluble compound limit was used to provide a guarantee of maximum safety) Concentrations exceeding 0 1 the MPC were recorded on only 7 occasions, two of which exceeded the MPC Both of these values did not exceed 0 1 of the MPC for insoluble ^{239}Pu compounds, however

3 *On days when maximum air sample values were found* the winds in the area had speeds of between 7 and 13 miles per hour

Since the weather can influence the behavior of plutonium in the soil it is interesting to compare rainfall wind velocity and temperature at Palomares with those at Rocky Flats This is tabulated as follows

	Palomares	Rocky Flats
Mean annual rainfall	7 9 inches	14 4 inches
Mean annual temperature	64 8°F	49 0°F
Velocity of most frequent strong wind gusts	43 mph	40 mph

Although Rocky Flats has a higher rainfall and a lower temperature the weather conditions are not extremely different

Thule

The accident at Thule was different from that at Palomares in that the bombs came down with the plane and were involved in a fire which was fueled by JP-4 jet fuel Also there was no soil involved just ice and snow

Contamination was spread over a drop-shaped area of 26 acres with the distribution in that area as shown in the following tabulation

DISTRIBUTION OF PLUTONIUM ON THE SURFACE IN THE VICINITY OF THE THULE CRASH[a]

Initial Plutonium Contamination $\mu Ci/100\ cm^2$	Initial Plutonium Contamination mCi/km^2	Area acres	Disposition
278	27776×10^3	0 49	Crust and packed
140	13969×10^3	2 23	snow removed to
9	912×10^3	3 43	an average
3	295×10^3	3 46	depth of
1	93×10^3	5 12	4 inches

[a] Excluding plutonium picked up on aircraft debris and that beyond blackened ice area

When fuel burned, a blackened area was produced on the snow Approximately 99% of the total plutonium found (3150 ± 630 g) was within this blackened area

This black crust contained unburned jet fuel It was estimated that as much as 18% of the fuel remained unburned Sedimentation studies showed that up to 80% of the plutonium was associated with low specific gravity debris that remained suspended in this jet fuel This debris included such things as metal, glass and nylon fibers, plastic, rubber, and flecks of paint The plutonium itself was in the form of oxide particles with a very wide size distribution

Core samples of the ice indicated that on the average 13% of the total plutonium was in the top 2 inches 36% in the top 4 inches and 45% in the top 6 inches About 15% was in the bottom 10 inches and the remaining 40% was distributed between 6 inches from the top and 10 inches from the bottom The total ice thickness was 34 inches The plutonium was distributed throughout the ice because the ice was fractured on impact of the plane with the ice It subsequently refroze It was estimated that there was a total of 350 grams of plutonium in the fractured ice area (approximately 0 5 acres)

A radiological survey made soon after the accident showed that most of the plutonium was confined to a limited area After the debris of the crash was removed another survey showed that the only significant plutonium contamination was confined to the snow and ice of the area where the fire had taken place

To determine if radioactivity had been spread via wind over large distances samples were taken from airplanes bound for Thule or passing across Greenland on ordinary traffic routes None of these samples showed activity above back ground The same was found for snow samples taken at places far from Thule

Contamination was removed by removing the contaminated snow – approximately 9000 cubic yards – and storing in empty steel fuel containers The radioactive water was returned to the United States

The amount of plutonium found in the ice was deemed low enough that dilution by melting would reduce its concentration to safe levels This melting was hastened by covering the area with ~~blank~~ black sand

7 RESULTS AND EVALUATION OF ROCKY FLATS PLUTONIUM SOIL SAMPLE ANALYSIS

Four Different Agencies Have Conducted Soil Sampling and Plutonium Analysis in the Soil Surrounding the Rocky Flats Site

Agency - 1 Colorado Committee on Environmental Information (CCEI)

Remarks

This is not a state sanctioned agency, but rather a group of interested citizens The actual work was performed by E A Martell and S E Poet of the National Center for Atmospheric Research Copies of a report by this group were mailed on January 13, 1970 to the following

E B Giller Director DMA-AEC Lloyd Joshel, General Manager, Dow Rocky Flats A R Tamplin, UCLRL Wright Langham, LASL, J H Harley, HASL R J Engelmann DEM-AEC H P Metzger, President, CCEI,

Later this report was released to the press and copies sent to the Governor of Colorado The data from that report are not inconsistent with other findings and were used in constructing ' models" of plutonium soil "contours ' constructed by Rocky Flats

Agency - 2 The Health and Safety Laboratory (HASL), USAEC

Remarks

These soil analyses data were consistent with the findings from this study by Rocky Flats The soil analysis data were used in constructing the "model' of plutonium soil contours A statistical analysis of the data from this report is found in Appendix B The HASL authors took considerable liberty in constructing their plutonium soil contours and also in making their estimates of total plutonium found in the soil The values for total plutonium calculated from the Rocky Flats study are somewhat lower than those estimated by HASL

Agency - 3 The Rocky Flats Health Physics Department

Remarks

Sampling and analysis specifically for plutonium in soil began in August 1969

Agency - 4 The Colorado Department of Public Health

Remarks

Their technique of taking a large number of samples from a large area and combining prior to analysis, rendered their data inappropriate for inclusion in the "model' constructed in this study However, analysis shows the data they reported are consistent with the findings of the Rocky Flats study

The first soil samples were taken in August of 1969 by The Rocky Flats Health Physics Department The sampling continued through June of 1970 A total of 99 sites extending as far as 10 kilometers (about 6 miles) from the plant were sampled In August of 1969 The Colorado Committee on Environmental Information under E A Martell took samples from about 18 sites Finally, in February of 1970 The Health and Safety Laboratory sampled and analyzed soil from 33 sites Only 18 of the 33 HASL samples were evaluated for this study The other 15 samples were taken beyond the Rocky Flats region and were not considered

The 135 soil sample sites were located by markers on a large contour map of Rocky Flats. The radial distance of each site from the barrel storage area was determined by measuring the distance between each marker on the map and the barrel storage area then using the scale division of the map As a result radial distances in kilometers were obtained to correspond to the soil sample analyses in mCi/km^2

The soil sample data were evaluated primarily to determine if specific levels of plutonium activity could be calculated It was also necessary to determine whether or not the soil sample data could be processed for meaningful results A theoretical basis for the study was a model constructed mathematically to represent observed data

Results

Figure 7 1 gives the first estimate of the dispersion of plutonium in soil over the Rocky Flats area and over land east of the Rocky Flats plant The isocurie contour lines give changes in plutonium activity continually from the 2000 mCi/km^2 level to the 13 mCi/km^2 level It can be seen that plutonium concentrations greater than 350 mCi/km^2 or 77 8 d/m/g dry soil (disintegrations per minute per gram of dry soil) cross the Rocky Flats boundary into private property

The isodose contour lines were constructed in the following manner First the contour map of Rocky Flats which contains all of the soil sample locations was divided into sectors The soil sample data for each sector were curve fitted using the method of least squares This resulted in a mathematical expression for each sector which gives the activity of the plutonium in the soil as a function of radial distance from the barrel storage area Specific levels of plutonium activity such as 2000 mCi/km^2 (444 4 d/m/g dry soil) followed by 1000 (222 2) 400 (88 9), 350 (77 8) 100 (22 2) 50 (11 1) and 20 mCi/km^2 (4 4 d/m/g dry soil) were selected The radial distance of each activity from the barrel storage area was then computed for each sector of the map An arc was struck across the sector at that distance and a center point was located on the arc The center points were then connected with a smooth but reasonably accurate curve for the level of activity desired

Additional Data

The most immediate result from having constructed the isocurie contour lines was the need for additional soil samples to be taken east of the Rocky Flats plant In December of 1970 after an interval of five months The Rocky Flats Health Physics Department sampled and analyzed soil from 38 new sites The samples were taken on private property between the Rocky Flats boundary and Indiana Avenue and on both sides of the access road leading to and from the plant Radial distances from the barrel storage area were determined and the new analytical data were incorporated into the model for isocurie contour lines As a result the isocurie contour lines were recalculated using 173 soil sample analyses Figure 7 2 shows the most accurate outline to date of plutonium in soil in and beyond Rocky Flats

From comparison to the isocurie contour lines in Figure 7 1 the outermost contours to the east and southeast appear not to have changed In contrast to the outer levels the regions which are described by levels of plutonium activity from 2000 mCi/km^2 to 350 mCi/km^2 (from 444 4 d/m/g dry soil to 77 8 d/m/g dry soil) show a slight easterly migration It is felt that the migration may be due to the gradual dissipation of plutonium contamination which is near the barrel storage area The area within the 2000 mCi/km^2 contour which shows activities greater than 2000 mCi/km^2 did not change significantly This may imply that the release of plutonium from the barrel storage area is now negligible It is also possible that the gradual dissipation of plutonium from within the 2000 mCi/km^2 contour may cause continued dispersion of plutonium at reduced activities particularly within the 1000 400 and 350 mCi/km^2 contours

In general the contours were less extensive in every direction except for two localized regions of plutonium activity The first has already been described (the 1000, 400 and 350 mCi/km^2 contours beyond the Rocky Flats boundary) The second is the protruding finger of plutonium activity in Sector 1 A of Figure 7 2 The activity resulted from a relatively high value reported in a soil sample taken near Walnut Creek just north of the plant The protruding contours did not change significantly from the initial study but appear exaggerated because the contours in the adjacent sectors are less extensive

To better define the present isodose contour lines additional soil samples will be taken on private property southeast of the Rocky Flats boundary toward Standley Lake and the upper and lower Twin Lakes Also soil samples will be taken just north of the Rocky Flats plant near Walnut Creek which is northeast of the plutonium process recovery complex on Atomic Energy Commission property

Quantities of Plutonium

The burden of plutonium in soil inside and outside the Rocky Flats boundary was determined mathematically by applying integral calculus to the equations used to project plutonium activity The total quantity of plutonium 239 dispersed in soil other than that contained by the asphalt pad was calculated to be 14 3 ± 2 0 grams The dispersion is over 8 35 km^2 of land (2063 acres) The quantity of plutonium-239 inside the Rocky Flats boundary is 6 7 ± 0 4 grams over 2 72 km^2 of land (672 acres) The amount of plutonium-239 on public and private property is 7 6 ± 1 8 grams over 5 63 km^2 of land (1391 acres) The quantities of plutonium were calculated by integrating the areas between the 2000 mCi/km^2 contour and the 13 mCi/km^2 contour The 13 mCi/km^2 contour was the

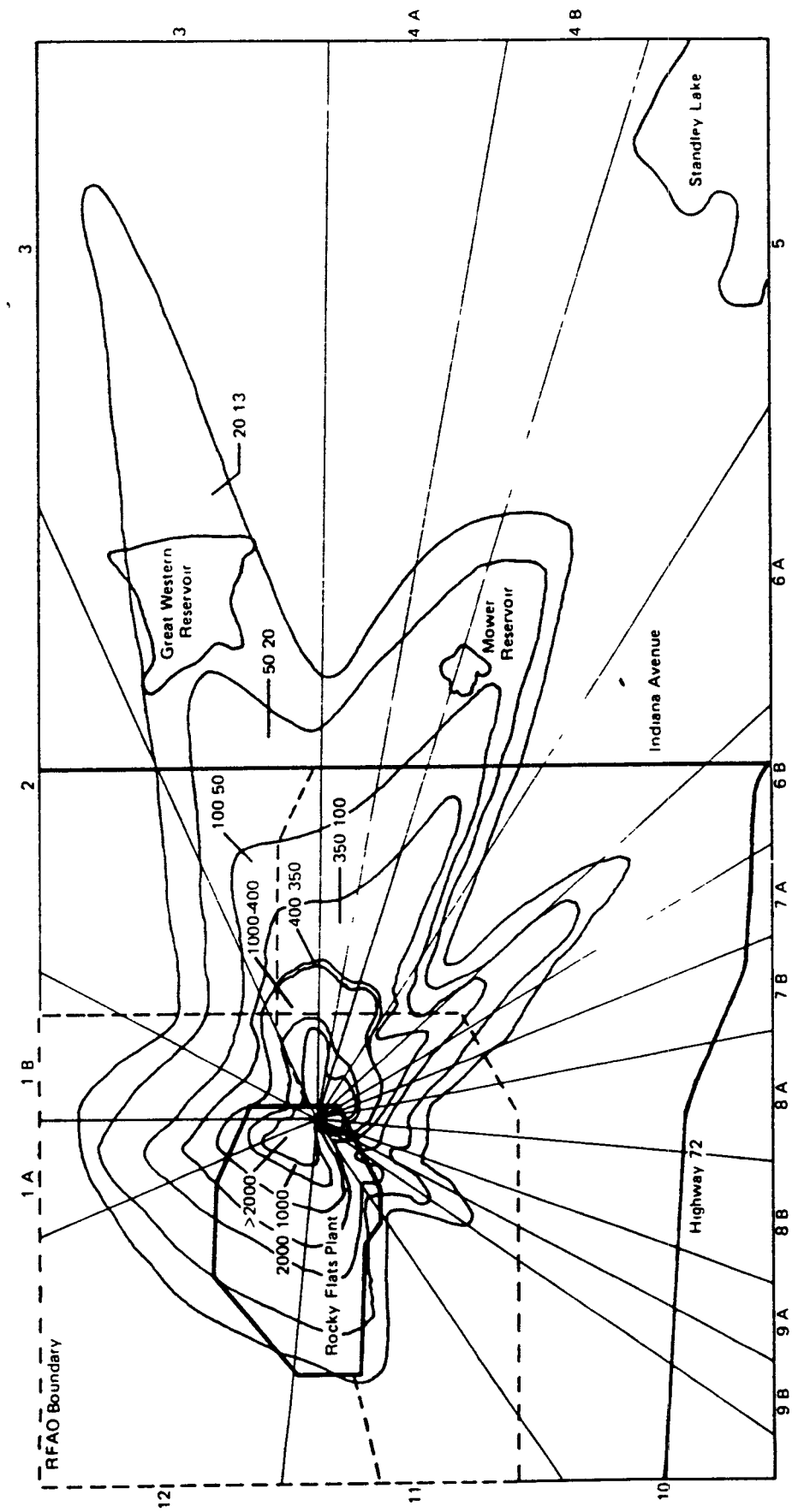


Figure 7 1 An Outline of Rocky Flats Showing the Levels of Plutonium Activity in the Soil in mCi/km² Soil Sample Data for the Contours were Evaluated per Sector of the Outline One Inch of the Outline is about 3600 Feet

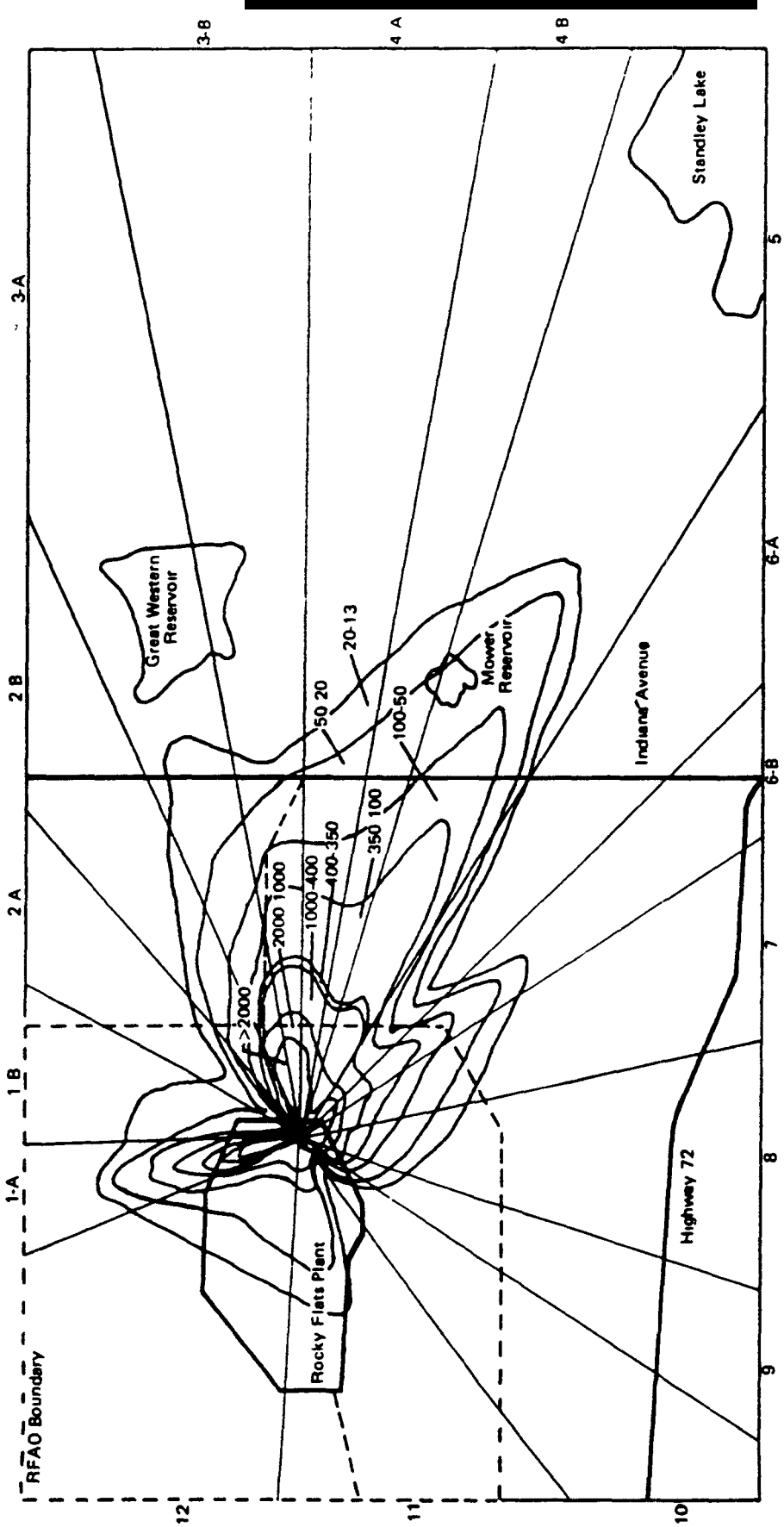


Figure 7.2 The Recalculated Isocurie Contour Lines Showing the Levels of Plutonium Activity in the Soil in mCi/km^2 Soil Sample Data for the Contour Lines were Evaluated per Sector of the Outline One Inch of the Outline is about 3600 Feet

minimum level considered Contours beyond this level gradually reach background which is about 1 mCi/km²

The amount of plutonium which was deposited on the soil of the barrel storage area is estimated to be about 85 grams Considering the maximum quantity of plutonium that may have been dispersed it is possible that about 80% of this amount of plutonium is still at the barrel storage area

A comparison of the quantities of Pu 239 per sector of the outline in Figure 7-2 shows that Sector 3 B has the greatest burden of plutonium per km² of land The value is 3 55 ±0 91 gms/km² Sector 1 A contains 3 12 ±0 94 gms/km²

As a final estimate of the quantity of plutonium which is dispersed in soil east of the Rocky Flats boundary the burden of plutonium per contour *outside* the boundary is given in Table 7-1

Table 7 1 Quantities of Plutonium Per Contour Outside the Rocky Flats Boundary *

Activity in mCi/km	Pu 239 in Grams	Area in km	Average** d/m/g Dry Soil
2000-1000	0 29 ± 0 02	0 02	198
1000-400	1 25 ± 0 09	0 13	131
400 350	0 32 ± 0 03	0 05	87
350 100	3 10 ± 0 5-	1 06	40
100-50	1 26 ± 0 4-	1 09	16
50 20	1 04 ± 0 5	1 98	7
20-13	0 34 ± 0 2-	1 30	4
13 1			- 2***
TOTAL	7 6 ± 2 0		

* In the calculations that convert the plutonium analytical data from d/m/μm to quantity of plutonium in grams it is assumed that we are looking at the pure plutonium 239 isotope Because of the distribution of isotopes in Rocky Flats plutonium the radiometric counting data will yield slightly higher values This assumption that we are making in the calculations of the total plutonium inventory will give a result in grams that is slightly higher than the actual gram-quantity found in the soil

** Assuming a density of dry soil of 1 gm/cm³ and a soil sample depth of 1 cm

*** Background plutonium activity is considered by the Rocky Flats Health Physics Department to be 0 2 d/m/g dry soil The Colorado Department of Health has on record a background activity of from 0 04 d/m/g to 0 11 d/m/g dry soil See Appendix B for an estimate of background using the HASL data

How the Dispersion of Plutonium in Soil was Estimated

The method of least squares[1] was used as a means to project plutonium activity given a set of soil sample data The technique consists of deriving a mathematical equation to express the soil sample results This is done by first selecting a general equation which gives the dosage d as a function of radial distance r The constants of the equation are solved using the data Then theoretical calculations are made and compared to the actual values of d The differences between the theoretical and experimental values are tabulated for a measure of how well the equation works

To demonstrate the effectiveness of the method consider the linear equation

$$d = a + br. \qquad (1)$$

We wish to use Equation (1) to curve fit a set of soil sample data From applying the method of least squares to Equation (1) normal equations in the form of

$$a \cdot n + b\Sigma r_i = \Sigma d_i$$

and

$$a\Sigma r_i + b\Sigma r_i^2 = \Sigma r_i d_i \qquad (2)$$

can be derived Here a and b are the constants to be solved r_i and d_i for $i = 1, 2, \ldots n$ are the radial distances in kilometers from the barrel storage area and the radioactive dosages in mCi/km² respectively and n is the number of samples Solving Equation (2) for the constant a results with

$$a = \frac{\Sigma r_i^2 \Sigma d_i - \Sigma r_i \Sigma r_i d_i}{n\Sigma r_i^2 - (\Sigma r_i)^2}$$

and for b

$$b = \frac{n\Sigma r_i d_i - \Sigma r_i \Sigma d_i}{n\Sigma r_i^2 - (\Sigma r_i)^2} .$$

Now Equation (1) becomes meaningful in that the constants have been determined from the various summations and products of the soil sample data Equation (1) can be used to project plutonium activity by simply computing the distance to correspond to some value d Linear functions however do not readily express soil sample results Therefore many possible equations including nonlinear functions which are solvable by the least-squares method were tested

Table 7 2 gives the equations which were programmed and used in a digital computer to curve fit soil sample data The results were immediate Four hyperbolic equations (those equations indicated by an asterisk) were selected by the computer to be the most effective equations By evaluating discrete sets of data, hyperbolic equations were readily derived for the dispersion of plutonium in soil Sets of soil sample data were obtained by dividing the soil sample

[1] D D McCracken and W S Dorn *Numerical Methods and FORTRAN Programming* John Wiley and Sons Inc New York 1964 p 262

map of Rocky Flats into sectors This limited the number of points in a set of data In this manner the formidable problem of evaluating over a hundred data points all at once was avoided Also as will be seen the use of map sectors made the integration for total quantities of plutonium quite simple

Map sectors were made by originating lines at the barrel storage area then extending the lines across the face of the map The size of a sector was made to depend upon the dispersion of the points and to more or less follow a specific direction e.g. Sector 1 is the northern sector If the number of points in a sector were too numerous or the data too poor to successfully determine a workable equation then the sector was subdivided into two parts and a second attempt at curve fitting the data was made The hyperbolic equations used to construct the isodose contour lines shown in Figure 7 2 are summarized in Table 7 3

Table 7 2 Equations Programmed to Evaluate Soil Sample Data

Function	Type	Results
$d=a+br$	linear	poor
$d=a+br^{1/2}$	parabolic	poor
$d^2=a+br$	parabolic	rejected
$d=a+br+cr^2$	parabolic	poor
$d=a+br^{1/3}$	cubical parabolic	poor
$d^3=a+br$	cubic	rejected
$d=a+b^{1/4}$	quartic	poor
$d^4=a+br$	quartic	rejected
$d=a+b\ \ln(r+1)$	logarithmic	poor
$d=a+b\ \ln(r)$	logarithmic	poor
$d=1/\lvert a+b\ \ln(r)\rvert$	inverse logarithmic	poor
$d=ae^{br}$	exponential	poor
$d=ae^{br^2}$	exponential	poor
$d=ae^{br}/r^2$	exponential	poor
$d=e^{(a+br+cr^2)}$	exponential	poor
$d=ae^{b/r}$	exponential	possible
$d=ae^{br}/r$	exponential	possible
$d=ae^{br^2}/r$	exponential	possible
$d=ar^b$	hyperbolic	possible
$d=a+b/r$ *	hyperbloic	usable
$d=a+b/r^2$ *	hyperbolic	usable
$d=a+b/r^3$ *	hyperbolic	usable
$d=a+b/r^4$ *	hyperbolic	usable
$d=1/(a+br)$	hyperbolic	poor
$d^2=a+br^2$	hyperbolic	rejected
$d=r/(b+ar)$	hyperbolic	poor
$d=r/(a+br+cr)^2$	–	poor

*The most effective equations

As can be seen from Table 7 3 the value of k in the general equation

$$d = a + \frac{b}{r^k}$$

is either 1 2 3 or 4 The value of k is data oriented to give the best results For example if soil samples which are taken near the barrel storage area show high concentrations of plutonium and the concentrations diminish rapidly with soil samples taken further away from the barrel storage area then the dispersion of plutonium can best be expressed by $k = 3$ or $k = 4$ On the other hand if the concentration diminishes slowly and is considerable say at two kilometers from the barrel storage area then $k = 1$ or $k = 2$ gives the best values of d Finally, if the dosage is so insignificant that d does not appear to increase or decrease over a given radial distance r then $k = 1$

Table 7 3 Plutonium Activity d as a Function of the Radial Distance r from the Barrel Storage Area per Sector of the Contour Map of Rocky Flats

Sector	Function	No of Points	General Direction of the Sector
1 A	$d=73/r^4-7$	5	N
1 B	$d=11/r+3$	7	N
2 A	$d=5/r+5$	12	NNE
2 B	$d=106/r-60$	7	NE
3 A	$d=133/r^4+11$	17	ENE
3 B	$d=689/r^3-32$	15	E
4 A	$d=451/r^3-6$	15	E
4 B	$d=275/r^2-9$	11	ESE
5	$d=463/r-95$	13	SE
6 A	$d=35/r^4-1$	6	SE
6 B	$d=133/r^2-25$	5	SSE
7	$d=23/r^3+4$	11	S
8	$d=7/r^3-3$	9	S
9	$d=14/r^4+6$	12	SSW
10	$d=1/r+$	8	W
11	$d=18/r-2$	7	W
12	$d=11/r+1$	13	NW

Considerable effort was spent in determining the value of k and hence, the hyperbolic equation to use for each sector of the map This is due to the fact that soil sample data are complicated and difficult to evaluate In all cases calculated values of d were compared to the actual soil sample results By assuming that high concentrations of plutonium are limited to a localized region and that the dispersion of plutonium in soil diminishes with greater distances from the barrel storage area it was possible to select the value of k based upon the nature of the data The activity of the barrel storage area which was estimated to be about 3.7×10^5 mCi/km^2 was also used as a point source to determine k

How the Effectiveness of the Curve Fitting Method Was Estimated

The additional soil samples provided by The Rocky Flats Health Physics Department were used to estimate the effectiveness of the curve fitting method This was done by comparing projected values of dose to the actual soil sample analyses Table 7-4 gives the results for all 38 samples It can be seen that some of the larger doses were underestimated by the model used previously while at the same time the smaller doses were overestimated The projected activity amounted to a 7 2% increase over the actual activity Using the additional soil sample analyses along with those obtained earlier the recalculated isodose contour lines now show an increase of only 3 2% over the additional soil samples

Table 7 4 The Comparison of Projected Plutonium Activity to Actual Soil Sample Results

Map Site No	Dose in mCi/km²	Projected Value*	Error	Value Using the Present Contours**
B-107	1397 2	769 8	-627 4	1050
B-115	772 2	406 5	-365 7	546
B-116	564 8	394 9	-169 9	362
B-110	414 4	418 8	4 4	380
B-108	369 4	769 8	400 4	703
B-109	324 4	368 4	44 0	338
B-105	262 4	341 9	79 5	181
B-117	252 9	244 3	-8 6	223
B-106	212 0	428 1	216 1	242
B-126	199 4	61 2	-138 2	56
B-113	127 8	201 9	74 1	92
B-118	112 0	332 7	220 7	301
B-121	103 0	100 5	-2 5	89
B-114	76 0	229 5	153 5	514
B-104	73 8	12 2	-61 6	41
B-125	56 7	50 7	-6 0	65
B-120	55 8	100 4	44 6	94
B-112	54 9	192 0	137 1	87
B 124	49 0	49 5	0 5	62
B-127	45 0	61 2	16 2	58
B-128	32 8	69 0	36 2	56
B-111	26 1	6 4	-19 7	27
B-129	21 6	72 8	51 2	63
B-130	11 7	27 8	16 1	14
B-131	11 4	28 9	17 5	14
B-132	11 1	30 2	19 1	14
B-133	11 0	30 9	19 9	15
B-138	10 9	46 0	35 1	38
B-134	10 8	31 8	21 0	18
B-136	10 8	26 3	15 5	27
B 137	10 8	25 9	15 1	26
B-135	10 7	26 8	16 1	27
B-123	8 6	53 2	44 6	22
B-119	7 6	81 5	73 9	32
B 102	7 2	5 4	-1 8	8
B-103	5 8	8 8	3 0	9
B 122	3 6	42 4	38 8	18
B-101	3 2	3 4	0 2	8

*Based on model using data obtained earlier

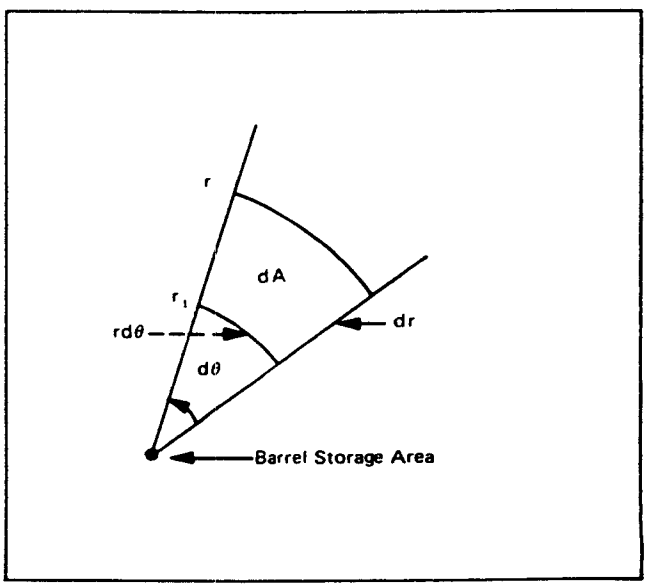
**This model uses the additional soil sample data along with that obtained earlier

In evaluating all of the soil sample data, which follow Table 7 6 an attempt to simulate the natural conditions which spread plutonium in soil was not made Wind direction and velocity were not considered, neither were natural washes in the foothills which dilute or accumulate plutonium concentrations The source of plutonium in the soil was assumed strictly to be the barrel storage area Other sources such as stack effluent, world-wide plutonium fallout, and the September 11, 1957 fire were not considered

How Quantities of Plutonium were Determined

The quantity of plutonium between two levels of plutonium activity was determined by integrating the area between the two levels The levels were drawn by first selecting a specific activity for each level with one level being closer to the barrel storage area than the other Figure 7-3 shows the two levels for any sector of the soil sample map of Rocky Flats The element of area dA between r_1 and r_2 is the area to be integrated Regarding the element of area dA as a rectangle its area will be the product of a pair of

Figure 7 3 The Element of Area dA in Polar Coordinates.



adjacent sides say dr and $rd\theta$ where $d\theta$ indicates the size of the sector. The result is

$$dA = rdrd\theta. \tag{3}$$

Integrating Equation (3) with respect to dr and $d\theta$ gives

$$A = \int_0^{\theta} \int_{r_1}^{r_2} rdrd\theta$$

or

$$A = \frac{\theta}{2} (r_2^2 - r_1^2), \tag{4}*$$

where θ = the angle between each side of the sector

The area A will be in units of km^2

To determine the quantity of plutonium over the area A the integrand of Equation (4) is modified to contain the hyperbolic function which gives dose as a function of radial distance r. It is stated as

$$Q = \int_0^{\theta} \int_{r_1}^{r_2} f(r,\theta)rdrd\theta, \tag{5}$$

where Q = the quantity of Pu 239 in millicuries. Using the actual hyperbolic equation, Equation (5) becomes

$$Q = \int_0^{\theta} \int_{r_1}^{r_2} \left(a + \frac{b}{r^2}\right) rdrd\theta$$

or

$$Q = \theta b \int_{r_1}^{r_2} \frac{rdr}{r^2} + \frac{\theta a}{2}\left[(r_2^2 - r_1^2)\right]. \tag{6}*$$

The area of each sector was integrated from the maximum plutonium activity of 2000 $mCi\ km^2$ to the minimum plutonium activity of 13 mCi/km^2. The partial quantities of plutonium were then summarized for the total dispersed over the area of the contours. In soil surrounding the Rocky Flats plant there are 14.3 grams of plutonium 239. The quantity, of course, extends from the 2000 $mCi\ km^2$ level. The area affected is 8.35 km^2 or about 2063 acres.

The integration was extended to determine the quantity of plutonium inside and outside the boundary of Rocky Flats. These calculations give the burden of plutonium in soil on public and private property. Figure 7-4 shows a sector being divided by the boundary.

The area inside the boundary was determined by subtracting the area of the shaded triangle from the element of area dA. The triangle was formed by striking an arc at the junction of the boundary and one side of the sector.

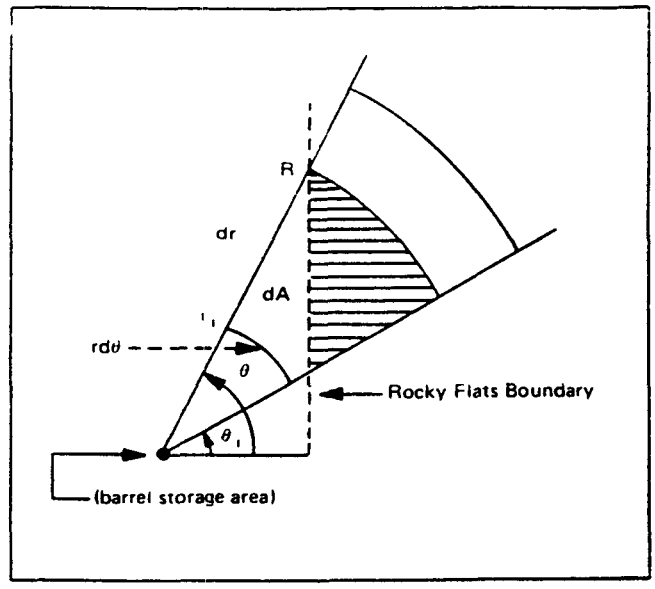


Figure 4 The Element of Area dA Divided by the Boundary of Rocky Flats

The area of the shaded triangle was determined from the calculus. The formula is

$$A_t = \int_{\theta_1}^{\theta_2} \int_{\frac{x}{\cos(\theta)}}^{R} rdrd\theta. \tag{7}$$

Integrating Equation (7) gives

$$A_t = \frac{R^2}{2}(\theta_2 - \theta_1) - \frac{x^2}{2}(\tan(\theta_2) - \tan(\theta_1)), \tag{8}*$$

where A_t = the area in km^2
R = the junction radius in kilometers,
x = the perpendicular distance of the Rocky Flats boundary in kilometers from the barrel storage area

By modifying Equation (7) the quantity of plutonium in the shaded triangle is determined by

$$Q_t = \int_{\theta_1}^{\theta_2} \int_{\frac{x}{\cos(\theta)}}^{R} \left(a + \frac{b}{r^2}\right) rdrd\theta$$

* Programmed for the digital computer

or

$$Q_t = \int_{\theta_1}^{\theta_2} \left[b \int_{\frac{x}{\cos(\theta)}}^{R} \frac{r}{r^k}\, dr + a \int_{\frac{x}{\cos(\theta)}}^{R} r\, dr \right] d\theta \qquad (9)^*$$

The junction radius R the angles θ_1 and θ_2 and the perpendicular distance x are all measured for a sector of the map Equation (9) therefore gives the quantity of plutonium in the described triangle of the sector By considering in the same manner each sector that is divided by the Rocky Flats boundary partial quantities of plutonium were determined inside and outside the boundary Finally the partial quantities were summarized The quantity of plutonium 239 inside the boundary of the Rocky Flats plant from the plutonium activity level of 2000 mCi/km^2 was calculated to be 6 7 grams The area of land affected is 2 72 km^2 which is about 672 acres Beyond the boundary on public and private property up to the 13 mCi/km^2 level there are 7 6 grams of plutonium over 5 63 km^2 of land which is about 1391 acres

Of particular interest is the quantity of plutonium within each specific contour east of the Rocky Flats boundary By using the element of area dA between individual levels and considering those segments of land that are divided by the boundary partial quantities of plutonium in each sector were summarized for the total amount of plutonium per contour Table 7 5 gives the results

Table 5 Quantities of Plutonium 239 Per Contour East of the Rocky Flats Boundary

Activity in mCi/km^2	239 Pu in Grams	Area in km^2
2000-1000	0 29	0 02
1000-400	1 25	0 13
400-350	0 32	0 05
350-100	3 10	1 06
100-50	1 26	1 09
50-20	1 04	1 98
20-13	0 34	1 30

How Probable Error was Computed

The calculation of probable error was taken as a necessary measure to determine the maximum and minimum quantities of plutonium in soil Probable error is determined by the variation in soil sample data and can be calculated directly from what is known as the normal law of error This concerns primarily the standard deviation of each of the least squares constants a and b of the general equation

$$d = a + \frac{b}{r^k} .$$

From the defining equation for probable error[2] the standard deviation S_a of the constant a is determined by

$$S_a = \left[\frac{\sum \frac{1}{r_i^{2k}}}{n \sum \frac{1}{r_i^{2k}} - \left(\sum \frac{1}{r_i^n} \right)^2} \right]^{\frac{1}{2}} \times \left[\frac{\sum \left(d_i - a - \frac{b}{r_i^k} \right)^2}{n - 2} \right]^{\frac{1}{2}} \qquad (10)^*$$

and the standard deviation S_b of the constant b is determined by

$$S_b = \left[\frac{n}{n \sum \frac{1}{r_i^{2k}} - \left(\sum \frac{1}{r_i^k} \right)^2} \right]^{\frac{1}{2}} \times \left[\frac{\sum \left(d_i - a - \frac{b}{r_i^k} \right)^2}{n - 2} \right]^{\frac{1}{2}} . \qquad (11)^*$$

Here r_i and d_i for $i = 1, 2 \ldots n$ data points are the data points in the set of data

[2] A G Worthing and J Geffner "Treatment of Experimental Data 8th ed John Wiley and Sons Inc New York 1959 p 249

* Programmed for the digital computer

The standard deviations S_a and S_b are combined to give the maximum and minimum quantities of plutonium in soil. The formula used is called the propagation of error equation and is given by

$$S^2(Q) = \left(\frac{\partial Q}{\partial a}\right)^2 S_a^2 + \left(\frac{\partial Q}{\partial b}\right)^2 S_b^2 \quad (12)^*$$

Here $S^2(Q)$ is the variance in the quantity of plutonium. The partial derivative terms $(\partial Q/\partial a)$ and $(\partial Q/\partial b)$ are solved by taking the partial derivatives of

$$Q = \theta b \int_{r_1}^{r_2} \frac{r\,dr}{r^n} + \theta a \int_{r_1}^{r_2} r\,dr$$

which is the expression for the quantity of plutonium in a given sector of the soil sample map. By the same token the variance $S^2(Q_t)$ in Q_t the quantity of plutonium in a described triangle is given by

$$S^2(Q_t) = \left(\frac{\partial(Q_t)}{\partial a}\right)^2 S_a^2 + \left(\frac{\partial(Q_t)}{\partial b}\right)^2 S_b^2 \quad (13)^*$$

where

$$Q_t = \int_{\theta_1}^{\theta_2} \left[b \int_{\frac{x}{\cos(\theta)}}^{R} \frac{r}{r^k}\,dr + a \int_{\frac{x}{\cos(\theta)}}^{R} r\,dr \right] d\theta.$$

Finally, the standard deviation S for some total quantity of plutonium e.g. the total quantity of plutonium outside the Rocky Flats boundary is determined by combining the variances of plutonium quantities in all of the sectors considered. This can be stated as follows:

$$S = \pm \left[S^2(Q_1) + S^2(Q_2) + S^2(Q_t) - \cdots \right]^{\frac{1}{2}}. \quad (14)^*$$

The calculations for probable error give the result $Q \pm S$ for quantities of plutonium. This means that inside the Rocky Flats boundary there are 6.7 ± 0.4 grams of plutonium-239. The burden of plutonium outside the Rocky Flats boundary on public and private property is 7.6 ± 1.8 grams. The total to be expected is 14.3 ± 2.0 grams of plutonium-239.

The mathematical methods used to determine probable error are based entirely on the soil sample data. No attempt was made to consolidate the systematic errors to be expected from the radiochemical analysis of the soil nor the error involved in determining the general location of each soil sample from the barrel storage area. For the calculations for probable error, the soil sample data were assumed to be unbiased and to follow a normal distribution about some mean.

Specific Activity, Density of Soil, and Conversion Factors

The specific activity of plutonium 239 was used extensively to (1) determine the activity of the barrel storage area, (2) convert dose in millicuries to total quantity in grams of plutonium 239, and (3) calculate quantities in grams of plutonium 239 to correspond to the levels of activity in mCi/km^2.

The specific activity of any radioactive isotope is defined as $g\,\lambda$ where g is the number of active nuclei per unit weight of the isotope and λ is the decay constant. The decay constant is given by 693/T where T is the α particle half life of the isotope. For plutonium 239 the specific activity is calculated to be

$$\text{Sp Act}_{239} = g \cdot \lambda = \left[\frac{6.02\times10^{23}}{239.052}\text{gms}\right] \times \left[\frac{.693}{2.436\times10^4\ \text{yrs}}\right]\left[\frac{\text{yr}}{3.156\times10^7\ \text{sec}}\right] = 2.270\times10^9 \frac{\text{dis}}{\text{gm sec}}.$$

* Programmed for the digital computer

The activity of the barrel storage area was determined and used as a point source to facilitate the evaluation of soil sample data From an estimate of the contaminated waste that was once stored in barrels over the area the amount of plutonium lost is considered to be about 85 grams Considering the major isotope (Pu 239) the activity of the barrel storage area in mCi/km^2 becomes

$$\text{Sp Act} = \left[\frac{2.27\times10^{9}\ \text{dis}}{\text{gm sec}}\right]\left[\frac{85\ \text{gms}}{380\times400\ \text{ft}^2}\right] \times \left[\frac{\text{mCi}}{3.7\times10^{7}\ \text{dis/sec}}\right]\left[\frac{\text{ft}^2}{9.29\times10^{-8}\ \text{km}^2}\right]$$

$$\simeq 3.7\times10^{5}\ \text{mCi/km}^2.$$

Quantities of plutonium 239 in millicuries can be converted to quantities in grams by the relationship 1 gram = 61 35 millicuries This is determined by

$$\text{Sp Act}_{239} = \left[\frac{2.27\times10^{9}\ \text{dis}}{\text{gm sec}}\right] \times \left[\frac{\text{mCi}}{3.7\times10^{7}\ \text{dis/sec}}\right]$$

$$= 61.35\ \text{mCi/gm Pu-239}.$$

Measured concentrations of plutonium in soil are given in units of d/m/g dry soil or disintegrations per minute per gram of dry soil To convert from d/m/g dry soil to units of activity per area or to millicuries per square kilometer the density of the soil and the depth of the soil sample must be considered The relationship is

$$1\ \text{d/m/g} = \left[\frac{1\ \text{dis}}{\text{gm}\cdot\text{min}}\right]\left[\frac{\text{mCi}}{3.7\times10^{7}\ \text{dis/sec}}\right] \times \left[\frac{\text{min}}{60\ \text{sec}}\right]\left[\frac{10^{10}\ \text{cm}^2}{\text{km}^2}\right] \rho h$$

or

$$1\ \text{d/m/g} = 4.5\ \rho h\ \text{mCi/km}^2$$

where ρ = the density of dry soil in gms/cm^3

and h = the depth of the soil sample in centimeters

To convert the soil sample analyses The Rocky Flats Health Physics Department used a density of 1 gm/cm^3 and a soil sample depth of 1 centimeter The additional soil samples obtained in December of 1970 were actually taken to a depth of 1 centimeter Previous soil samples, however, were taken to depths ranging from 3 to 5 centimeters These samples were obtained between August of 1969 and June of 1970

The Colorado Committee on Environmental Information used a density of 1 gm/cm^3 and a soil sample depth of 1 centimeter Their soil samples were also taken to a depth of 1 centimeter, but in August of 1969

Finally The Health and Safety Laboratory USAEC used a density of 1 2 gm/cm^3 down to a depth of 15 centimeters and a density of 2 4 gm/cm^3 below 15 centimeters For the most part, the soil samples were taken to a depth of 20 centimeters [3] Appropriate calculations were made for the changes in soil sample depth The soil samples were obtained in February of 1970

The accumulation of plutonium is affected by its vertical distribution in soil For the purpose of converting data in this study and except for the HASL soil samples, the dispersion of plutonium was assumed to be within the first centimeter of topsoil

To demonstrate the trace quantities of plutonium that are associated with the levels of activity used in the study, the levels of activity were converted to grams of plutonium using the specific activity of plutonium-239, a soil sample density of 1 gm/cm^3 and a soil sample depth of 1 centimeter The results are given in Table 7-6

Table 6 Specific Activity in Grams of Plutonium-239

mCi/km^2	d/m/g dry soil	Pu-239/gram of dry soil
2000	444 4	$3\ 26\times10^{-9}$
1000	222 2	$1\ 63\times10^{-9}$
400	88 9	$6\ 54\times10^{-10}$
350	77 8	$5\ 72\times10^{-10}$
100	22 2	$1\ 63\times10^{-10}$
50	11 1	$8\ 16\times10^{-11}$
20	4 4	$3\ 24\times10^{-11}$

[3] P W Krey and E. P Hardy "Plutonium in Soil Around The Rocky Flats Plant HASL-235, The Health and Safety Laboratory USAEC, New York 1 August 1970

Soil Sample Data

The Rocky Flats Health Physics Results of Soil Samples Taken August 1969 - June 1970 on Atomic Energy Commission Property

Map Site	Pu Activity in mCi/km^2	Pu Activity in d/m/g Dry Soil
G 1	20 7	4 6
G 2	33 3	7 4
G 3	2 7	0 6
G 4	13 1	2 9
G 5	17 6	3 9
G 6	270 9	60 2
G 7	9 0	2 0
G-9	2 7	0 6
G 10	18 0	4 0
G-11	9 5	2 1
G 12	5 0	1 1
G 13	14 9	3 3
G 14	373 5	83 0
G 15	2128 5	473 0
G 18	5 4	1 2
G-19	35 1	7 8
G 20	1377 0	306 0
G 22	1431 0	318 0
G 23	48 6	10 8
G 25	396 0	88 0
G 26	6 3	1 4
G 27	8 1	1 8
G 28	39 6	8 8
G 29	117 0	26 0
G 30	7 2	1 6
G 31	8 6	1 9
G 32	26 1	5 8
G 33	10 8	2 4
G 34	9 0	2 0
G 35	9 0	2 0
G 36	5 9	1 3
G 37	3 2	0 7
G 38	0 9	0 2
B 18	6 3	1 4
B 19	6 3	1 4
B 20	10 4	2 3
B 21	1 3	0 3
B 22	6 3	1 4
B 23	6 8	1 5
B 24	6 8	1 5
B 30	5 4	1 2
B 32	5 4	1 2
B 33	69 8	15 5
B 34	16 7	3 7

The Rocky Flats Health Physics Results of Soil Samples Taken August 1969 - June 1970 on Public and Private Property within a Six Mile Radius of the Rocky Flats Plant

Map Site	Pu Activity in mCi/km^2	Pu Activity in d/m/g Dry Soil
B 1	3 8	0 8
B 2	1 1	0 2
B 3	4 5	1 0
B 4	4 3	0 6
B 6	2 3	0 5
B 7	4 1	0 9
B 8	0 5	0 1
B 9	5 0	1 1
B 10	1 8	0 4
B 12	9 0	2 0
B-13	1 8	0 4
B-14	0 5	0 1
B-15	11 7	2 6
B-16	4 3	1 0
B-17	27 9	6 2
B 25	2 3	0 5
B-26	9 5	2 1
B-27	11 3	2 5
B-28	5 4	1 2
B-29	5 0	1 1
B-35	2 3	0 5
B-36	0 5	0 1
B-37	3 2	0 7
B 38	3 6	0 8
B-39	8 1	1 8
B 40	3 6	0 8
B 41	0 5	0 1
B-42	4 1	0 9
B-43	9 9	2 2
B 44	2 7	0 6
B 45	12 2	2 7
B 46	0 5	0 1
B 47	5 4	1 2
B-48	3 6	0 8
B-49	5 9	1 3
B 50	3 6	0 8
B 51	9 5	2 1
B 52	17 6	3 9
B 53	6 3	1 4
B-54	3 2	0 7
B-55	3 6	0 8
B-56	37 8	8 4
B-57	4 1	0 9
B 58	20 7	4 6
B 59	3 6	0 8
B-60	5 0	1 1
B 61	9 0	2 0
B-62	20 7	4 6
B 64	14 9	3 3
B-65	5 0	1 1
B-66	5 9	1 3
B-67	6 8	1 5
B-68	6 3	1 4
B-71	4 1	0 9
B 79	0 9	0 2

The Rocky Flats Health Physics Results of Additional Soil Samples Taken in December 1970 on Private Property east of the Rocky Flats Plant

Map Site	Pu Activity in mCi/km	Pu Activity in d/m/g Dry Soil
B-107	1397 2	310 5
B 115	772 2	171 6
B-116	564 8	125 5
B 110	414 4	92 1
B-108	369 4	82 1
B 109	324 4	72 1
B-105	262 4	58 3
B-117	252 9	56 2
B-106	212 0	47 1
B-126	199 4	44 3
B-113	127 8	28 4
B-118	112 0	24 9
B 121	103 0	22 9
B 114	76 0	16 9
B-104	73 8	16 4
B 127	56 7	12 6
B 120	55 8	12 4
B 112	54 9	12 2
B 124	49 0	10 9
B 127	45 0	10 0
B 128	32 8	7 3
B 111	26 1	5 8
B 129	21 6	4 8
B 130	11 7	2 6
B 131	11 4	2 5
B 132	11 1	2 5
B 133	11 0	2 4
B 138	10 9	2 4
B 134	10 8	2 4
B 136	10 8	2 4
B 137	10 8	2 4
B 135	10 7	2 4
B 123	8 6	1 9
B-119	7 6	1 7
B 103	7 2	1 6
B-122	3 6	0 8
B 101	3 2	0 7

The Colorado Committee on Environmental Information Results of Soil Samples Taken in August of 1969 on Public and Private Property within a Seven-Mile Radius of The Rocky Flats Plant

Map Site	Pu Activity in mCi/km²	Pu Activity in d/m/g Dry Soil
A	5 8	1 3
B	60 8	13 5
C	0 4	0 1
D	0 6	0 1
E	1 3	0 3
F	1 3	0 3
G	1 4	0 3
H	0 6	0 1
I	7 7	1 7
J	5 2	1 2
K	4 0	0 9
L	0 5	0 1
M	1 7	0 4
U	1 5	0 3
V	2 4	0 5
W	0 2	<0 1
X	0 8	0 2
Y	1 9	0 4

The USAEC Health and Safety Laboratory Results of Soil Samples Taken in February of 1970 on Public and Private Property within a Seven-Mile Radius of the Rocky Flats Plant

Map Site	Pu Activity in mCi/km²	Pu Activity in d/m/g Dry Soil
R 1	2 4	<0 1
R 2*	3 1	<0 1
R 3	4 2	<0 1
R 4	11 0	0 1
R 5*	15 0	0 1
R 6*	1950 0	16 0
R 7*	480 0	3 2
R 8*	630 0	–
R 9	2 6	<0 1
R 11	5 4	<0 1
R 12	47 0	2 1
R 13	50 0	0 6
R 14	17 0	<0 1
R 15	18 0	0 1
R 17	14 0	0 1
R 18	2 0	<0 1
R 19	8 0	<0 1
R 21	2 7	<0 1

* Samples taken on Atomic Energy Commission Property

8 CURRENT RADIATION LEVELS IN THE OIL STORAGE AREA

A gross gamma survey was performed on the entire surface of the asphalt pad Data from this survey were converted from relative radiation intensities into integers and trans posed to a graphic scale for evaluation Based on the results obtained by the gamma survey, four areas were selected for excavation soil sampling and analysis of the samples for radioactive contaminants

For the γ ray mapping four 4-inch by 2 inch NaI(Tl) de tectors arranged in a horizontal array on 18-inch centers were suspended from the rear of an IHC Scout With detector faces approximately 3 inches from the ground, the detector output was monitored and recorded via a ratemeter strip-chart recorder and the vehicle traveled at a slow and constant speed The entire Pad was scanned in 6 foot increments with a 9 to 10- inch overlap on each pass

The strip-chart recorder data were reduced by dividing the relative readings into integers from one to twenty-five and transposing these integers to a scale graph The relative gross-gamma profile thus obtained is shown in Figure 8 1 The numbers represent only the relative gamma-ray readings at the pad surface Each integer increment on the figure represents a change in counting rate of 1 to 2% As seen in the figure several highly localized areas (shown in red) were found which showed γ-readings significantly above the general background Two of these labeled 17 21 and '25 in Figure 8 1 were deemed hot spots that is showed activity levels >15% above the general background Another significant feature of the profile is the large areas of similar activity levels over the entire pad

Additional verification of these gamma activity levels was provided by a survey performed by P H Dodd and R F Droullard of the AEC s Geophysical Branch from the Grand Junction Office Utilizing gamma ray analy sis equipment in a mobile laboratory designed for uranium ore evaluation and modified for soil monitoring this survey made measurements at the "hot spots,' at other locations on the pad off the pad and north across the street The measurements indicated activity levels at the hot spots to be at least twice those elsewhere on the pad or off the pad Attempts to identify the source of the anoma lies were partly successful Ratios of two gamma ray energy ranges (1 00 to 2 80 MeV and 0 600 to 1 00 MeV) were measured at several sites The results showed a ratio lower by a factor of approximately 2 at the hot spots This is interpreted to mean a slightly higher concentration of either ^{239}Pu or ^{235}U is contributing a higher percentage of low energy gamma rays to the natural activity levels

Table 8 1 Lab Analyses of Test Hole Samples

Sample	Wt of Sample (grams)	Total Uranium	U [illegible]	Wt U (kg)
14 22	7294	0 6	<0 01	0 043
14 23	7065	0 9	<0.01	0 06
14 24	6893	28	0 12	1 93
14 25	6503	17	0 07	1 1
14 26	7334	42	0 15	3 1
14 27	7850	43	0 15	3 8
14 28	7797	41	0 14	3 2
14 29	8242	41	0 14	3 4
14 30	7056	34	0 11	2 4
14 31	7520	26	0 08	1 9
14 32	7450	35	0 15	2 6
14 33	7162	20	0 09	1 4
				~25 kg
17 11	7805	0 8	<0 01	0 06
17 12	7978	18	0 07	1 4
17 13	7750	26	0 10	2 0
17 14	7585	10	0 04	0 8
17 15	7369	6	0 02	0 4
17 16	7097	5	0 01	0 3
17 17	7677	7	0 02	0 5
17 18	7323	3	<0 01	0 2
17 19	6949	3	<0 01	0 2
				~6 kg

Sample	Wt of Sample (grams)	Plutonium ppb	Wt Pu (μg)
SW 5	8378	0 01	0 084
SW 10	7833	36	282
SW 15	7578	70	530
SW 20	6373	34	216
SW 24	7620	2 5	19
			~1047 μg
NC 4	7809	0 003	0 023
NC 7	7385	0 033	0 24
NC 8	7395	0 74	5 5
NC 10	7391	11	81
NC 13	6582	0 89	5 8
			~92 6 μg

To quantitatively evaluate the measurements taken on the pad similar gross gamma readings were taken at the pad and at several other site locations For these a single 4 inch by 2 inch NaI(Tl) crystal was placed on the surface and the counts recorded on a scaler-timer unit The new parking lot east of 111 Building, the 750 Building parking lot and the asphalt in front of the 881 guard post gave similar readings of 13,000 to 16 000 counts per minute Furthermore, the asphalt in all areas including the pad counted 25 to 30% higher than nearby shoulder or gravel areas Pad readings were in the range of 15,000 to 16,000 counts per minute with the exception of areas near

[redacted]

Figure 8-1 Results Of Gamma Survey Of Asphalt Barrier Showing Relative Values (See text for explanation)

[redacted]

and on the hot spots Therefore the gamma profile of the pad indicates a general level of activity similar to other asphalt areas That the asphalt appears to be 25-30% hotter than nearby gravel surfaces may be due to a concentration of natural uranium and/or its daughters which occurs during the asphalt production process It is known that the front range shows a high natural uranium content in the soil Also it is noteworthy that highways offsite in this area show γ-ray readings similar to those found on site Thus with the exception of the 'hot spots' on the pad it appears that a sensitive measurement of the asphalt thickness may have been performed

Following the γ-ray mapping, the two "hot spots were γ-ray pulse-height analyzed in an effort to determine whether or not the activity detected was emanating from ^{239}Pu or natural/enriched uranium These efforts were unsuccessful If plutonium or uranium were spilled on the original ground layer the γ ray emissions would have to penetrate 6 inches to 8 inches of gravel fill plus 3 inches to 5 inches of asphalt to be detected The low energy plutonium γ rays are severely attenuated A simple experiment was performed to see if high-energy gamma rays could be used to distinguish between uranium and plutonium Spectra were taken using (NaI(Tl) detectors of natural uranium enriched uranium and ~5 year-old WR plutonium sources with 4-inch and 8-inch concrete slabs interposed between the sources and detector The 5-year-old plutonium source was chosen to simulate the average age of plutonium released in the drum-storage area There were no readily distinguishable differences in the spectra

It was then decided by the Committee to excavate four selected areas Areas No 14 and No 17 represent the two 'hot spots labeled '17 21 and '25 " respectively in Figure 8-1 Hole SW an area thought to be a likely spot for plutonium, and Hole NC a possible "background' area The areas (2- to 4-sq ft) were covered with a tent the asphalt removed and holes excavated Material removed was placed in 1-gallon polyethylene bottles and sent to Building 881 Analytical Laboratories for analysis Digging was continued until the NaI(Tl) and/or alpha monitors indicated background levels The holes were refilled with new fill material and resealed with asphalt

The results of the wet chemical analyses are summarized in in Table 8-1 Holes No 14 and No 17 contained large amounts of depleted uranium Based upon these percentages we removed approximately 25 kg of depleted uranium from No 14 and 6 kg from No 17 A sample from each hole was γ-ray scanned with a Ge(Li) detector and no evidence for plutonium was seen Holes No SW and No NC conversely showed no uranium but did show detectable amounts of plutonium The total plutonium removed from holes No SW and No NC was estimated from five analyses and therefore should be considered as only an order of magnitude number These values are ~10 mg for No SW and ~200-300 μg for No NC

In the summary the following statements can be made concerning the pad survey

a The γ ray mapping indeed detected areas of activity above "background" levels Two areas of significant activity were located

b The γ-mapping further showed large general patterns of similar activity These patterns may indicate activity on the old ground layer or may merely be a measure of the natural uranium content, and thus the thickness of the asphalt layer

c Two of the localized activity areas which were excavated, No 14 and No 17, resulted from depleted uranium This was further evidenced by the presence of a very concentrated depleted uranium contaminated oil layer at 18 in and 30 in depth for holes No 17 and No 14, respectively (Table 8-2)

d Analysis of the four test holes (Table 8-1) showed little or no mixing between Pu and U Thus, reasonably rigid segregation of barrels must have been maintained during the 'lifetime of the storage area

e In no case was activity found to be migrating upwards from the original ground level into the fill material

f While activity in holes No 14 and No SW extended several inches into the soil (Table 8-2), in all four cases no activity was found more than 1 in into the clay layer Thus the clay layer, which varies in depth from 4 inches to 15 inches below the original ground level, appears to serve as a natural barrier to the further vertical migration of the radioactive material

Table 8 2 Summary of Results

Hole	Activity From γ Map Thru Asphalt (cpm)	Maximum γ Reading in Hole (cpm)	Maximum α Reading in Hole (dpm)	Active Material	Where Activity Firs Detected	Vertical Thickness of Activity Layer (inches)	Location of Clay Layer (below top of pad) (inches)
No 32	55 000	3×10^6	5000	Depleted U	original ground layer	6 8	30
SW	16 000	<16 000	20 000	Pu	original ground layer	4 6	16
No 17	35 000	2×10^6	5000	Depleted U	original ground layer	1 2	18
NC	16 000	20 000	1000	Pu	original ground layer	<1	26

9 SOIL STABILIZATION

In order to have an additional tool available during any future contamination control and removal operations and to specifically assist in minimizing resuspension during the removal of the soil under the asphalt pad a study of soil stabilizers in Rocky Flats soil was initiated

Several methods exist for stabilizing fine-grained particles of soil or dust which are potential sources of air and water pollution Soil stabilization can be accomplished by physical chemical and vegetative methods or by combinations thereof

Stabilization by chemical and/or combined chemical and vegetative means can serve several useful purposes at Rocky Flats The following are some possible uses

1 Immobilization of contaminated soil which might result from a 'spill'

2 Prevention of contaminated soil dispersion during removal of the asphalt pad

3 Enhance the establishment of permanent vegetation in and around Rocky Flats

4 Reduce soil erosion and subsequent property damage by airborne debris during periods of high velocity winds

Evaluation of soil stabilizers has been in progress for approximately one year Preliminary results[1] suggested that J-197 a product commercially available from Dowell Division Dow Chemical U S A showed some degree of effectiveness in stabilizing the soil when applied at a concentration between 60 and 100 pounds per acre It was recommended that further evaluation of J-197 should be performed

A survey of available literature[2-6] showed that many chemicals have been evaluated by the United States Bureau of Mines and others In addition to J-197, two soil stabilizing products (Coherex® and Peneprime®) cited in the literature are being evaluated at Rocky Flats Methods of application have been developed and stability tests have been underway for approximately 9 months [7]

Following preliminary evaluation of the three soil stabilizing chemicals combined chemical and vegetative soil stabilizing experiments[8] are in progress The following is a review of the results obtained during evaluation of chemical soil stabilizers and the results of recent combined chemical and vegetative soil stabilization research at Rocky Flats

General information describing the three soil stabilizers of interest is given in Table 9 1 Evaluation of these chemicals has been conducted in test plots of varying sizes all within the general vicinity of the asphalt pad (903 storage area) Figures 9-1 and 9-2 show the location of six plots utilized to develop application methods and to evaluate the stabilizing qualities of each chemical These plots were established between October and December 1970

Table 9-1 General Information On Three Soil Stabilizing Chemicals.

Product Name	Manufacturer	General Description of Product
Coherex®	Golden Bear Division Witco Chemical Co	Non-volatile emulsion consisting of 60% semi liquid natural petroleum products and 40% wetting agents
J 197©	Dowell Division Dow Chemical U S A	Poly acrylamide a plastic material similar to surfactant chemicals and thickening agents contains 1% violet dye for marking purposes
Peneprime®	Empire Petroleum Co	Asphalt derivative of petroleum crude bottoms

J 197 was applied at concentrations varying from 17 to 75 pounds per acre (test plots 1 2, 3, and 6) A water solution of Coherex was applied to test plot number 4 (1 part Coherex to 4 parts water) at a concentration of 0 5 gallons per square yard Test plot number 5 was stabilized with Peneprime (1 gallon per square yard)

To gain more experience on the feasibility of stabilizing large areas of soil with varying surface features, a plot (number 6) about 0 9 acre in size was stabilized Surface features included coarse gravel fine soil, and grassy areas A total of 7,500 gallons of J-197 solution was applied with a portable pump and fire hose The concentration of J-197 powder was 70 pounds per acre

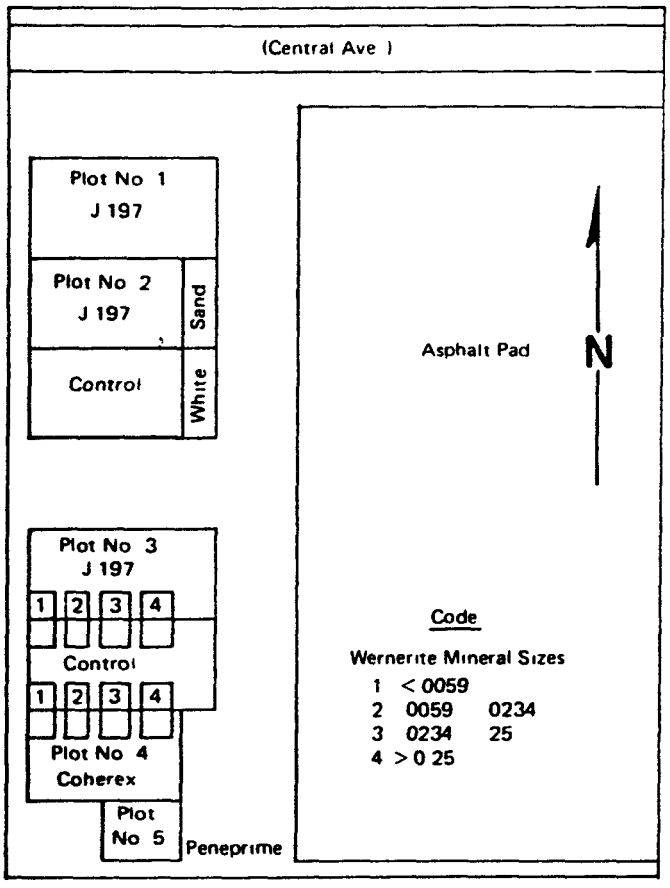


Figure 9-1 Soil Stabilization Test Plot Locations.

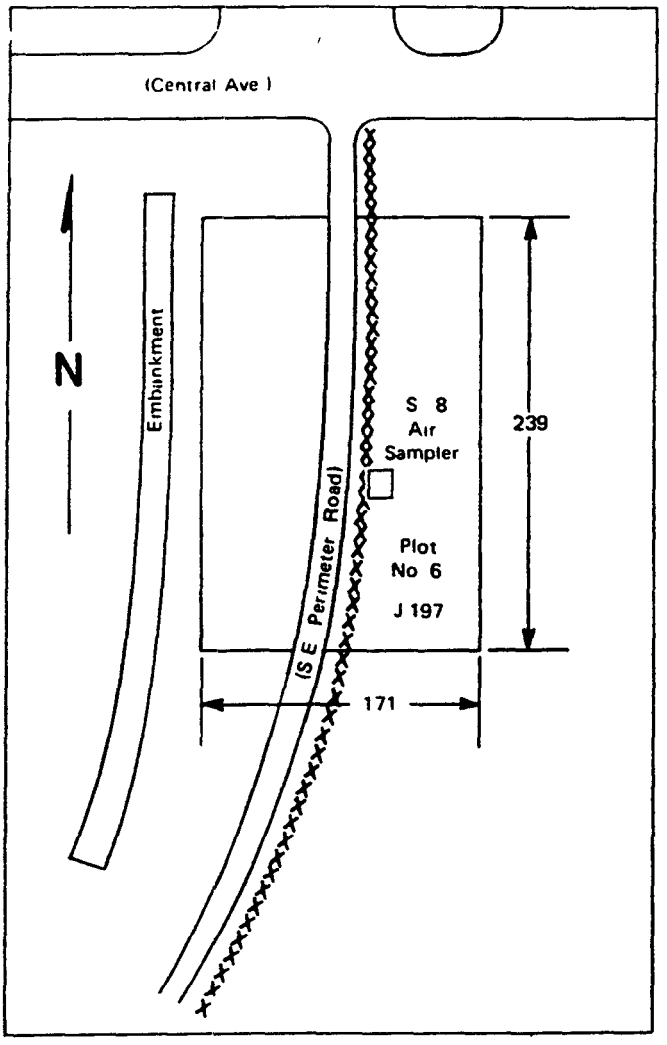


Figure 9 2 Soil Stabilization Test Plot Locations

Test plot number 6 surrounds a Health Physics air sampler (number S-8) This air sampler has consistently indicated relatively higher amounts of airborne radioactive contaminants when compared to other air samplers located along the east perimeter fence Therefore, data from this air sampler might be used to evaluate the effectiveness of J-197 on this test plot

Criteria for evaluation of each chemical soil stabilizer consisted of

1 Ease of solution preparation and distribution

2 Visual observation

3 Depth of penetration

4 Ability to prevent particle dispersion (test plots 2 3 and 4)

5 Effectiveness on various soil types (coarse gravel to fine soil)

6 Permanence (weather resistance)

7 Ecological and aesthetic considerations

8 Effect on established vegetation

9 Evaluation of air sample data (test plot number 6 only)

Evaluation of particle dispersion prevention (criteria number 3) was conducted by using an underlay of white sand (test plot number 2) which could be visually observed Wernerite, a fluorescent mineral, was used as an underlay on test plot numbers 3 and 4 It was also used on the common edges of the intervening control plot Four different particle sizes were used Erosion of this mineral was followed by observation with the aid of ultraviolet light

Visual inspection of test plot number 2 (stabilized with J 197) indicated temporary stability The white sand remained in place somewhat longer than the sand on the adjacent unstabilized control plot However the stabilized sand did dissipate within two months The method of application and total concentration (17 pounds of J 197/acre) of stabilizer applied was far from optimum The J 197 was applied manually which accounts for the low concentration Before further work was conducted a method of spraying the J-197 solution was developed This method employs a portable pump fire hose and adjustable nozzle to distribute the solution (see Figures 9-3 and 9-4)

Evaluation of test plots number 3 and 4 indicates that both J-197 and Coherex have been fairly successful in retaining the fluorescent mineral The mineral on these plots was examined on March 30 1971 (using ultraviolet light) After 4½ months of exposure to the high winds and precipitation some erosion of the smaller mineral particles from the plot stabilized with J-197 was evident but considerable more loss occurred on the control plot Table 9 2 compares the mineral retention on these two test plots and the control plot

Table 9-2 Wernerite Mineral Retention On Two Stabilized Test Plots And A Control Plot (a)

Mineral Size (in)	J 197 Test Plot No 3	Coherex Test Plot No 4	Unstabilized Control Plot
< 0059	~20%	~90%	~10%
0059 - 0234	~50%	~90%	~20%
0234 - 25	~80%	~95%	~60%
- 25	95%	95%	95%

(a) The percents given are estimates based on visual examination and serve only as a relative guide for comparison of J 197 and Coherex stabilizing products

Test plot number 5 was effectively stabilized with Peneprime However as described previously (see Table 9-1) this chemical is an asphalt product and is undesirable from several standpoints It is difficult to apply, detrimental to established vegetation and has no aesthetic appeal

The airborne contamination data obtained from the air sampler located in the center of test plot number 6 (S-8 air sampler) did not show any significant change It appears that the test plot was not large enough to have an effect S-8 air sampler data from November 12, 1970, to the present can be reviewed in Figure 3-1 in Section 3 Visual examination of this plot of S-8 air sampling data coupled with knowledge of the wind pattern and velocity and knowledge of activity in the area indicated that J 197 will stabilize all types of Rocky Flats soil A crust of varying thickness is formed when this chemical is applied Also relatively large particles will be held in place by the crust Figures 9 5 9 6 and 9 7 show the type crust formed in gravel fill soil and grassy areas This protective crust was found to be friable and will deteriorate in time Therefore it appears that this stabilizer would be excellent for short term stabilization but semiannual application may be necessary if long term stability is required

Evaluation of J 197 Coherex and Peneprime as chemical stabilizers is summarized in Table 9-3 Based on these results J 197 and Coherex were considered favorable for further testing

Table 9 3 Evaluation Of Three Soil Stabilizing Chemicals

Information	J 197	Coherex	Peneprime
Solution preparation and distribution	Excellent	Excellent	Warm weather only (>70°F)
Visual Observations	Forms crust of varying thickness	Soil appears to be undisturbed after application	Similar to asphalt
Depth of Penetration	Approximately 1/32 to 1/2 inch	Approximately 1/8 to 3/4 inch	Approximately 1/2 inch
Prevention of Particle Dispersion	Good to Excellent	Good to Excellent	Excellent
Effectiveness on various soil types	Good on gravel and top soil	Good on gravel and top soil	Only test was on gravel fill excellent
Weather resistance	Crust is friable semi annual application may be necessary if long term stabilization is needed	Unaffected by weather	Unaffected by weather
Ecological and aesthetic considerations	Excellent	Good	Poor
Effect on Established Vegetation	No effect	No effect	Covers vegetation completely
Overall Rating	Good	Good	Poor

In May 1971 combined chemical and vegetative test plots were established in an area just east of the asphalt pad The area consisted of several inches of gravel fill material To provide a good base for vegetation an overlay of 3 inches of local top soil was added Three plots 8100 square feet in size were marked off Each plot was cross fertilized (350 pounds/acre), cross dragged, cross seeded with Fairway crested wheat grass (25 pounds/acre) and final cross dragged

Chemical stabilization was performed on two of the plots using solutions of Coherex and J-197 The third plot was used as a control plot for comparative evaluation A total of 2,000 gallons of J-197 solution was applied The resulting concentration was 110 pounds of J-197 per acre The Coherex solution was applied at a rate of 5 gallon per square yard A total of 450 gallons of solution consisting of 1 gallon of Coherex per 4 gallons of water was used The control plot was treated with 2000 gallons of water

Preliminary results of the test plots stabilized by combined chemical and vegetative means are very encouraging The final preparation of the plots was completed on May 4 and 5, 1971 (see Figure 9-8) In less than two weeks, seedlings were observed on both of the stabilized plots and the control plot Since that time, the growth of vegetation has been excellent Figure 9-9 shows the condition of the three plots one month after seeding The grass was about 3 inches tall on each plot with fairly uniform coverage Neither the Coherex nor J-197 were found to be detrimental to the establishment of vegetation If high winds occur, little soil erosion and subsequent loss of vegetation is expected from the stabilized plots

The results of soil stabilization research show that both Coherex and J-197 can be used for chemical and/or combined chemical and vegetative stabilization Coherex appears to be more favorable for use on sand or gravel surfaces However, J-197 is believed to be somewhat better for stabilization of top soil and grassy areas Both chemicals can easily be prepared and distributed on all types of terrains Water sprinkling trucks portable pumps and perhaps even aerial application methods can be employed

The cost of stabilization, including materials and labor, will vary considerably depending on the area to be treated It is estimated that the two products evaluated will be competitive in cost and will range from $200 to $400 per acre

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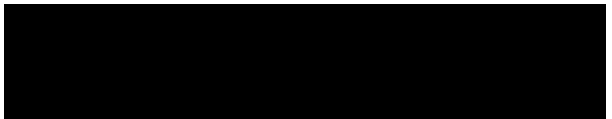


Figure 9-3 Preparation of Dowell J-197®Solution

[redacted]

Figure 9-4 Application of Dowell J 197® Solution To Seeded Test Plot

[redacted]

[redacted]

Figure 9 5 Crust From Gravel Test Plot Stabilized With Dowell I 197® Note Size Of Rock Held In Crust

[redacted]

[redacted]

Figure 9 6 Crust From Fill Soil Test Plot Stabilized With Drywall [illegible]

[redacted]

[redacted]

Figure 9-7 Crust From Fine Soil And Grassy Area

[redacted]

[redacted]

Figure 9-8 View Of Test Plots 10 Days After Stabilization By Combined Chemical And Vegetative Methods (Dowell J 197@foreground Coburn background)

[redacted]

[redacted]

Figure 9-9 View Of Test Plots 41 Days After Stabilization B, Combined Chemical And Vegetative Methods (Dowell J 197@left Coherex right)

[redacted]

10 POSSIBLE OPTIONS FOR DISPOSITION OF PLUTONIUM-CONTAMINATED SOIL

When considering the ultimate disposition of plutonium contaminated soil, the following actions were evaluated

1 Chemical recovery of plutonium from soil

2 Chemical mining or *in situ* leaching

3 Soil removal and burial at an approved site

1 Chemical recovery of plutonium from the soil would be an enormous task Concentrated acids at room temperature have been shown to remove between 49 and 72% in 1 hour The time can be reduced to less than one-half by processing at the solution boiling point This amount represents the plutonium held on the surface of the soil All the plutonium can be recovered by complete decomposition of the soil in an acid solution consisting of 15 8 *M* HNO_3 and >0 1 *M* HF With this method dissolution is completed in about one-half hour at the solution boiling point These recovery rates are based on laboratory studies and would not be expected on a large system Much intermediate waste would be generated by chemical removal processes This does not seem practical

2 Chemical mining appears to be very difficult because of the thin large area and the varying plutonium concentration In addition this type of processing is not considered to be very efficient Solubilized plutonium could be carried into the water-courses and eventually to humans This does not seem practical

3 One possible burial location for Rocky Flats contaminated soil may be the salt mine near Lyons Kansas However this site will not be ready to receive contaminated material until at least 1975 Also, the contamination level of Rocky Flats soil is so low that the soil may not be acceptable for burial

Commitments have been made by Dow (Rocky Flats) and the AEC to the Governor of Colorado that we would remove the "Pad" and the contaminated soil it entraps This can be accomplished whenever an approved AEC burial site can receive the material The cost associated with removal is estimated in the next section of this report (See Section 11) Decision on actions on additional contaminated soil such as the action of plowing that was used in Spain, cannot be made without a better plutonium concentration-depth profile The mechanism of plutonium transport in soil should be established before actions such as plowing can be considered

Soil stabilization studies were initiated and will be continued Data resulting from studies to this period are reported in Section 9 of this report

11 ESTIMATE OF THE COST OF REMOVAL OF PLUTONIUM CONTAMINATED SOIL UNDER THE ASPHALT "PAD"

Present information does not establish the exact depth or area from which the contaminated soil will be removed It is possible to estimate unit costs of labor packaging materials freight and burial or storage as follows

Unit	Cost per 55-Gallon Drum
Labor to Fill and Load Drums	$10 00
Packaging Materials	10 50
Freight - Based on 600 Pounds Per Drum and Freight Rate of $2 10/100 Pounds	12 60
Burial of Storage - Based on 7 4 Cubic Feet per Drum and Present NRTS Charges of $1 06/ Cubic Foot for Storage of Trans-uranium Wastes	7 85
TOTAL	$40 95 ~ $41 00

Using this information and the following assumptions regarding area and depth of contamination it is possible to indicate the magnitude of the operation

Case I

1 Assume the removal of the asphalt pad as uncontaminated waste Remove the 10 inches of fill dirt and 8 inches of contaminated soil under the asphalt pad as contaminated waste	220,000 Cubic Feet
2 Remove 4 inches of fill dirt and 2 inches of contaminated soil from the area east of the asphalt pad to the security fence as contaminated waste	111,500 Cubic Feet
TOTAL	331,500 Cubic Feet
331,500 Cubic Feet / 7 4 Cubic Feet/Drum	~47,360 Drums
47 360 Drums X $41 00/Drum	~$1,950,000
Equipment Facilities	~ 50,000
TOTAL	~$2,000,000

Case II

1 Assume the removal of the asphalt pad as uncontaminated waste Remove 10 inches of fill dirt and 24 inches of contaminated soil under the asphalt pad as contaminated waste	414,000 Cubic Feet
2 Remove 4 inches of fill dirt and 8 inches of contaminated soil from the area east of the asphalt pad to the security fence as contaminated waste	223,000 Cubic Feet
TOTAL	637,000 Cubic Feet
637 000 Cubic Feet / 7 4 Cubic Feet/Drum	~91,000 Drums
91,000 Drums X $41 00/Drum	~$3,731,000
Equipment and Facilities	~ 50,000
TOTAL	~$3,781,000

12 ROCKY FLATS CURRENT HEALTH PHYSICS ENVIRONMENTAL SAMPLING PLAN

Vegetation Samples

ON SITE

Vegetation samples are collected at 6-month intervals from 16 locations within the security fence The samples are currently analyzed for total U + Pu activity (gross alpha) and specifically for plutonium

OFF SITE

Sixty-seven locations with a 10-mile radius of the plant are collected at 6-month intervals All vegetation samples are collected from public right-of-way and confined to that normally consumed by grazing animals The samples are analyzed for gross alpha and specifically for plutonium

Water Samples

ON SITE

Water samples from holding ponds 1 5 and 9 (release points) are collected daily The samples are composited weekly and analyzed for uranium plutonium and americium

Monthly water samples from each of 14 sampling wells are analyzed for uranium and plutonium

OFF SITE

Thirty-seven samples from lakes and streams surrounding the plant are collected at 6-month intervals The samples are analyzed for gross alpha content A plutonium determination is made if the sample activity exceeds 0 7 pCi/liter ($0\ 7 \times 10^{-9}$ μCi/ml)

Samples from four reservoirs and nine community tapwater supplies are collected bimonthly and analyzed for total uranium plus plutonium activity In addition, these reservoir samples are analyzed specifically for plutonium Samples from Great Western (Broomfield water supply) and Standley (Westminster water supply) Reservoirs are analyzed specifically for americium

A weekly water sample collected from Walnut Creek at Indiana Avenue is analyzed for total uranium plus plutonium activity and specifically for plutonium and americium

Air Samples

ON SITE

Twelve on site continuous air samples are collected daily (except weekends and holidays) and analyzed for total long-lived alpha activity An analysis is also performed for beryllium

Five weekly, 6-hour, high-volume air samples taken east of the 903 Area are collected and analyzed for plutonium content

OFF SITE

Twelve high-volume, continuous air samples taken approximately 2 miles from the plant boundary are collected daily (except weekends and holidays), composited weekly, and analyzed specifically for plutonium content In addition, a 6-hour high-volume air sample is collected weekly at each of Wagner site (located approximately 2 5 miles ESE of the plant) and Coal Creek Canyon These air samples are analyzed specifically for plutonium content

Biweekly air samples from nine additional locations are analyzed for total long-lived alpha activity Samples from these locations are also analyzed for beryllium

Fallout (Dustfall) Samples

ON SITE

Five dustfall samples are collected bimonthly from locations on site and downwind from the production buildings One site is downwind from the 903 Area Fallout (dustfall) samples are analyzed specifically for plutonium

OFF SITE

Twelve dustfall samplers are located off site atop the air sample stations The samples are collected bimonthly

More remote samples are collected monthly from Berthoud and Castle Rock All dustfall samples are analyzed for plutonium content

Soil Sampling

The Rocky Flats Health Physics department maintains an extensive, routine soil sampling program Soil samples are collected from locations both on and off the plant site

Analysis of soil samples is generally confined to plutonium however a capability for analysis of uranium and americium also exists An analysis for other than plutonium would be performed based on historical sample results and/or mode of contamination

ON SITE

1 903 Area

Forty-eight samples are collected at 100-, 150-, and 500-ft distances from the nearest edge of the asphalt pad This area is presently being sampled four times per year It is planned to reduce this to twice per year in September 1971

2 Other Sites Within the Security Fence

Twenty-nine locations on alternate grid points of the 500 ft " Austin Company grid" are sampled twice per year and analyzed specifically for plutonium

3 Samples Between the Security and Cattle Fences

Sixty locations (predominantly east and south of the plant proper but covering all areas) are sampled semiannually

OFF SITE

1 Grid Samples

There are 20 locations on each of three concentric circles of 1 2 and 5 mile distances from the plant center These samples are approximately equi-spaced and are collected twice per year

In order to intensify the soil sampling in areas east and south of the plant boundary, 30 additional locations in these areas are sampled along public right-of way These samples are collected twice per year

2 Remote Off Site Samples

Twenty soil samples from Arvada, Westminster, and Denver locations between Boulder and Fort Collins, Coal Creek Canyon, locations between Leyden and Golden and along 104th Avenue are sampled twice per year

Sediment Samples

ON SITE

Sediment samples from each of the holding ponds are collected monthly for analysis of plutonium and americium This schedule will continue until any problems are adequately defined The schedule will then be reduced to twice per year

Sediment samples from Walnut Creek and Woman Creek effluent water courses are collected monthly These sediment samples are analyzed for plutonium and americium content

OFF SITE

Sediment samples from each of four reservoirs (Great Western Baseline Standley and Ralston) are collected twice each year and analyzed for plutonium An analysis specifically for americium is performed on Great Western and Standley Reservoir sediment samples

Chemical Contaminants

ON SITE

Water samples from holding ponds 1, 5 and 9 are collected three times per week and analyzed for pH, NO_3^-, PO_4^{-3}, F^- and total solids A daily water sample from the holding pond 5 is composited and analyzed for biological oxygen demand (BOD) These chemical analyses assure that all water leaving the plant is in compliance with presently accepted drinking water standards

Water from the 14 sampling wells is analyzed for pH NO_3^- PO_4^{-3} F^- and total solids

OFF SITE

Because of a recurrent problem with nitrates leaching into Walnut Creek from the solar evaporation ponds, a weekly water sample from Walnut Creek at Indiana Avenue is collected and analyzed for NO_3^- concentration During periods of peak spring runoff, a daily water sample from Walnut Creek at Indiana Avenue is analyzed for NO_3^- concentration

Presently, environmental sample analysis for plutonium and americium is restricted due to the lack of adequate alpha pulse-height analysis instrumentation This condition will be relieved with the purchase of additional instrumentation for the performance of the analyses in Fiscal Years 1972 and 1973

13 CONTINUING RESEARCH SUPPORT AT ROCKY FLATS ON THE STUDY OF PLUTONIUM IN SOIL

This study pointed out several areas where information is needed if reasonable decisions are to be made pertaining to problems of plutonium in soil The following activities are being or will be pursued at Rocky Flats if sufficient funding is available

1 Sampling and analytical techniques will be improved

2 Standard samples will be generated and stored for future use

3 Sampling exchange programs with other laboratories will be continued

4 Plutonium soil-depth profiles will be obtained

5 The mechanism of plutonium transport in soil will be studied

6 The chemical form of the plutonium in Rocky Flats soil should be identified

7 The solubility of the plutonium in Rocky Flats soil will be studied

8 The particle size of the plutonium species and the size of aggregate particles of plutonium and soil should be established

9 Emergency procedures to "fix" plutonium in soil for easy removal, in event of any future accidents, will be established

10 New and improved air sampling systems will be devised and deployed that specifically measure resuspended plutonium particles from Rocky Flats soil

11 The significance of multiple resuspension of plutonium particles from Rocky Flats Soil will be established

PART II RECOMMENDATIONS

1 It is recommended that the AEC purchase additional property surrounding the Rocky Flats Plant site

2 It is recommended that areas in which the soil contains plutonium in excess of 350 mCi/km^2 be fenced and restricted from grazing

3 It is recommended that additional soil stabilization action be pursued on soil in and around the Rocky Flats plant where plutonium concentration exceeds 350 mCi/km^2

4 It is recommended that additional vegetative cover be considered for use in conjunction with soil stabilizers in the areas east and south of the drum storage area

5 It is recommended that the asphalt "Pad," which is an effective seal, not be disturbed until all questions of a disposal site for the contaminated soil have been resolved

6 It is recommended that specific responsibility be assigned for ultimate disposal of the soil under the pad

7 It is recommended that the diagnostic research and health physics support described in Part I (13) of this report be completed

8 It is recommended that close liaison be maintained with other AEC and commercial agencies or sites using plutonium so that all new plutonium environmental information can be shared

PART III APPENDICES

Appendix A Geology of Rocky Flats Plantsite

Appendix B Analysis of HASL Data

Appendix C Soil Contamination and Asphalt Pad

Appendix D Simplified Conversion Scale and Table for the Various Units Used in the Literature to Express the Levels of Plutonium Contamination in Soil

Appendix E Trip Report

[redacted]

GEOLOGY OF ROCKY FLATS PLANTSITE

C T Illsley, December 1970

The Rocky Flats Plant in Colorado is located on a gently sloping terrace or outwash plain at the eastern edge of the foothills of the Front Range of the Rocky Mountains. Rock outcrops are extremely scarce in the immediate vicinity of the site and most of the geologic descriptions are based on extrapolations from adjacent areas to the north and south. Some information was obtained from surface rock outcrops west of the area and subsurface conditions were observed in the abandoned Capitol Mine (coal) which is only one half mile west of the plant. Geologic information was obtained from the maps and reports referred to in the list of references.

The surface soil is composed of 10 to 25 feet of alluvium, or gravel which consists mostly of quartzite boulders and clay. This surficial material is underlain by 700-800 feet of Laramie formation according to Spencer (1961) and shown in Figure A 1. Earlier reports indicated the presence of 25 to 30 feet of the Arapahoe formation but Spencer could not distinguish such a lithologic break in the Laramie formation in this area.

The Laramie formation is divisible into two parts. The lower part about 100 feet thick is composed chiefly of sandstone and sandy shale interbedded with lesser amounts

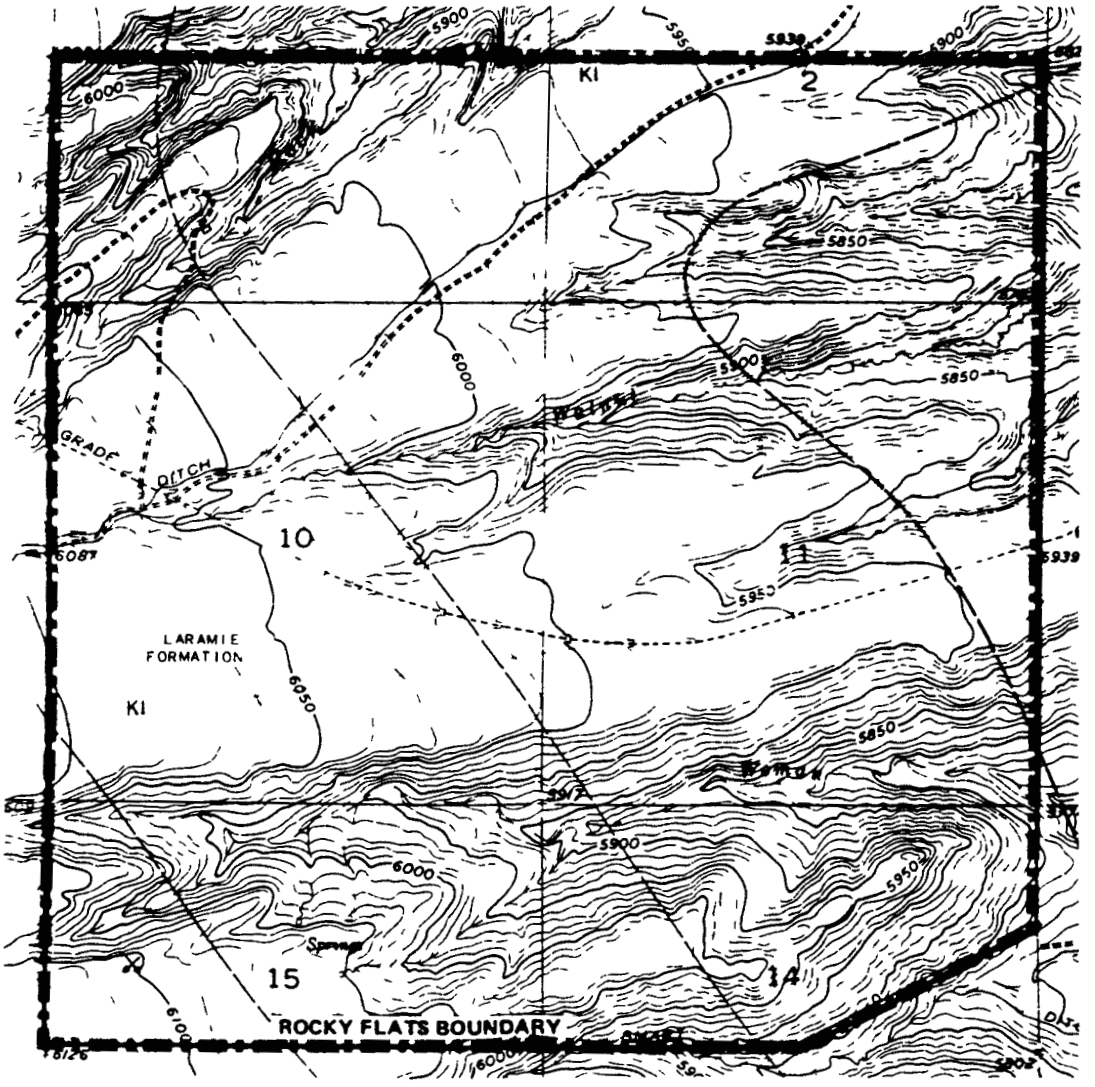


Figure A 1 Geologic Map of Rocky Flats Plantsite

[redacted]

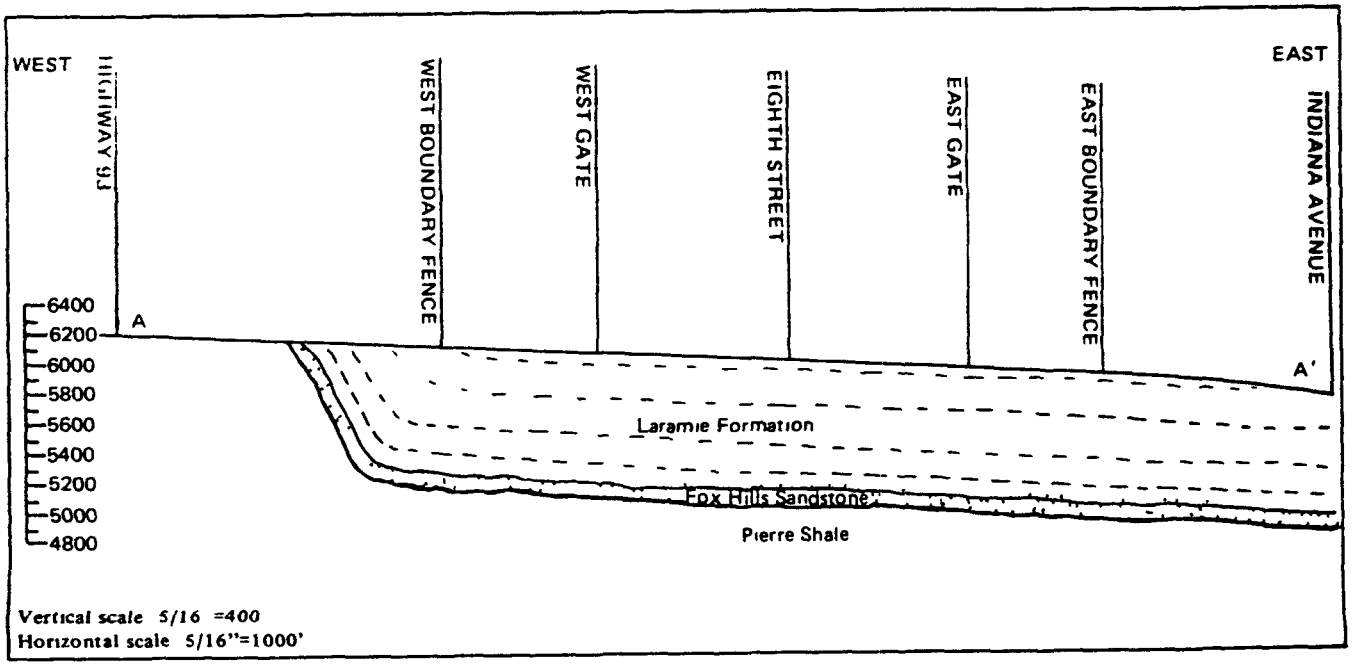
of clay fire clay shale and coal The upper part about 600 to 700 feet thick is composed chiefly of clay shale and sandy shale and some lenticular beds of sandstone and lignite

The Fox Hills sandstone underlies the Laramie formation The Fox Hills is a massive cross-bedded and ripple-marked sandstone that is conformable with the underlying Pierre shale The lower two-thirds of the Fox Hills is a fine-to-coarse-grained, slightly calcareous, yellow-to-greenish buff sandstone The upper one-third of the Fox Hills is a fine to-medium-grained light-gray to light-yellow mottled cross bedded sandstone The thickness of the Fox Hills sandstone varies from 60 to 250 feet between Ralston Creek and Superior beneath Rocky Flats site it is probably about 100 feet thick

The underlying Pierre shale is about 8000 feet thick It is a lead-gray to brown and black shale of marine origin Although generally homogeneous, the Pierre also contains some siltstone silty sandstone beds of limestone, and limestone concretions

In contrast to the steeply dipping rock strata west of the plantsite the structure beneath the plantsite is almost level and rather uncomplicated A cross section drawn along an east-west line from Indiana Avenue to Colorado High way 93 and based on data (from Spencer 1961) of structure contours on the top of the Fox Hills sandstone shows the generalized structure (See Figure A 2) The steeply dipping (~44° east) Fox Hills sandstone exposed on the surface one half mile east of Highway 93 is assumed to flatten out to less than 5° at a point close to the west gate of the plant It is logical to assume that the overlying conformable Laramie formation also is relatively flat under the plant site The structure is significant from a hydrology viewpoint because steeply dipping strata would tend to restrict the ground water to great depths below the surface and extending many miles east of Rocky Flats Since the layered rocks are actually almost level, water entering the ground water system from the Rocky Flats area would be more likely to reappear at the surface only a short distance downslope from the recharge area, such as in the drainage patterns of Woman Creek or Walnut Creek An illustration of possible ground water movement is shown in Figure A-3, a cross section along a southwest-northeast line through the area Evidence for such water migration is the presence of several intermittently flowing springs along the leading edge of the Rocky Flats pediments As indicated in Figure A-3 surface water entering the ground in the vicinity of 771 Building will likely emerge to the surface in Walnut Creek, possibly in the Rocky Flats holding pond If the water were to percolate deeper, it should still reappear west of Indiana Avenue To migrate east of Indiana the ground water would have to penetrate more than 350 feet of the Laramie formation This would be unlikely because of the impermeable nature of the clay-shale strata Faulting could create passageways for downward flow of ground water, but no evidence of faulting has been reported within the immediate area of Rocky Flats

Figure A 2 Geologic Cross Section of Rocky Flats Plantsite along Line AA



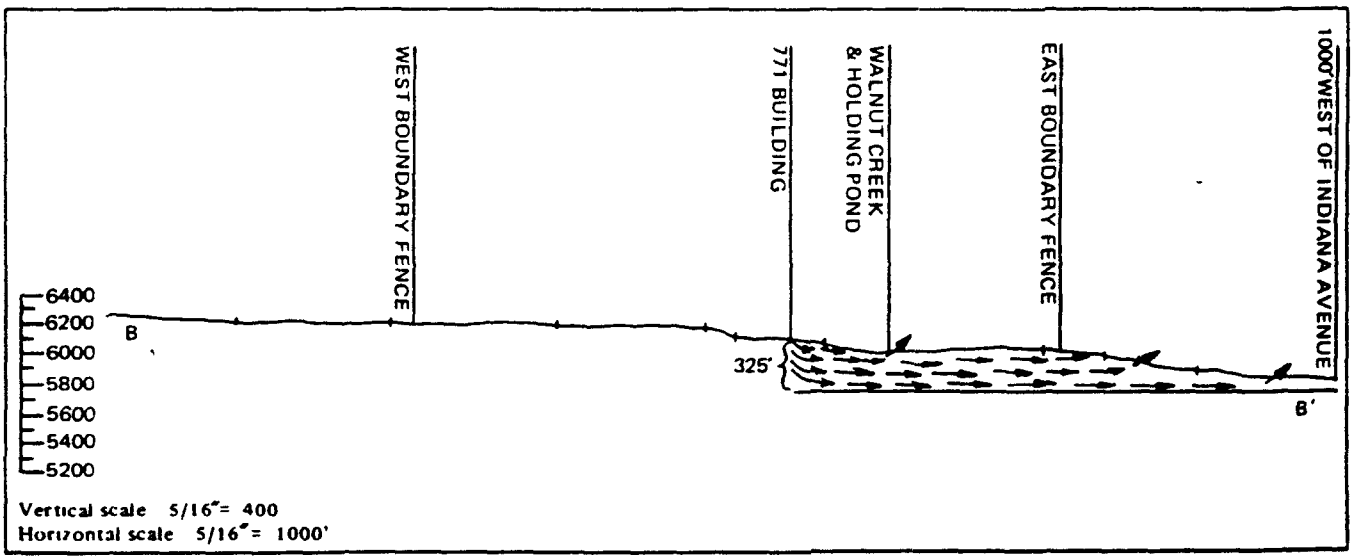


Figure A 3 Cross Section of Rocky Flats Plantsite along Line BB

References

1 F D Spencer 1961 *Bedrock Geology of the Louisville Quadrangle Colorado* U S Geol Survey Geol Quad Map GQ 151

2 R Van Horn 1957 *Bedrock Geology of the Golden Quadrangle Colorado* U S Geol Survey Geol Quad Map GQ 103

3 T S Lovering and E N Goddard 1950 *Geology and Ore Deposits of the Front Range Colorado* U S Geol Survey Prof Paper 223

4 C R Butler 1950 *Structure of Post Cambrian Formations in the Vicinity of Coal Creek Colorado* MS Thesis, University of Colorado

5 O W Hampton 1957, *Structural Geology of the Foothills Region from Plainview to Golden Colorado* MS Thesis, University of Colorado

6 F T Quiett 1951 *The Geology of the Plainview Area Jefferson County Colorado* MS Thesis Colorado School of Mines

ANALYSIS OF HASL DATA

Section 1

LOG-NORMAL ANALYSIS OF PLUTONIUM DATA

D E Michels

Log-normal analysis is a technique based on statistical considerations of how analytical values may vary from one another Data are plotted on probability paper such that the linearity of the array of plotted points yields statistical conclusions This technique applies to plutonium data and three kinds of conclusions result (1) When the array of plotted points is precisely linear the data are called homogeneous That is, the plutonium in all samples of the group is overwhelmingly from a single source, (2) The analytical value associated with the 50th percentile of the data is the average value for the group, (3) The standard deviation for the group is given by the slope of the array of plotted points The conclusions described above depend on finding straight-line plots when the data are plotted on probability paper Since required straight lines are obtained only when a logarithm scale is used for the analytical values (and not when a linear scale is used) the distributions are termed log-normal Log-normal distributions are generally obtained for a wide variety of trace materials studied by geochemists

This technique was applied to data reported by Health and Safety Laboratory (HASL 235) personnel since their 33 sample sites included some sites which would be expected not to include detectable amounts of plutonium from Rocky Flats Thus the HASL data involve two kinds of samples one kind dominated by world-wide fallout, the other kind dominated by Rocky Flats effluent The plot on probability paper permits a sharp distinction to be made between the two groups In addition, an independent estimate is obtained for the plutonium background in Denver soils that is due to world-wide fallout

Figure B-1 shows the probability plot for the HASL data which in Table B-1 are ranked in order of analytical values

The junction of the two components of the plot, at about 3 0 mCi/km^2, is the natural division between the background distribution of plutonium and the plutonium derived from Rocky Flats On the basis of Figure B-1, the HASL data are divided into two sub-groups which are replotted independently in Figure B-2 Each sub-group gives a statistically satisfactory fit to a straight line and we therefore conclude that each subgroup is homogeneous

The higher content subgroup is clearly caused by Rocky Flats since the samples which comprise the subgroup were taken near and downwind of the plant Although it is tempting to select an "average" value for this group by correspondence with the 50th percentile, that temptation should be resisted The anomaly has definite structural features which complicate the meaning of average value

However the lower-content subgroup if it truly is background should not have a complicating structure Hence this log-normal analyses yields an average value of 2 4 mCi/km^2 associated with the 50th percentile and is a valid measure of the Denver background

Table B 1 Order and Percentiles for Plutonium in Soil Samples

Sample	mCi/km^2	Percentile	Sample	mCi/km^2	Percentile	Sample	mCi/km^2	Percentile
33	1 8	3 0	9	2 6	36 4	4	11	69 8
18	2 0	6 1	20	2 6	39 4	17	14	72 8
27	2 0	9 1	24	2 6	42 5	5	15	75 8
29	2 0	12 1	21	2 7	45 5	14	17	78 8
31	2 1	15 2	32	2 7	48 5	15	18	81 8
10	2 2	18 2	30	2 8	51 5	16	19	84 9
22	2 2	21 2	23	3 0	54 5	12	47	87 9
26	2 3	24 2	2	3 1	57 6	13	50	91 0
1	2 4	27 3	3	4 2	60 6	7	480	94 0
25	2 4	30 3	11	5 4	63 6	8	630	97 0
28	2 5	33 4	19	8 0	66 7	6	1950	–

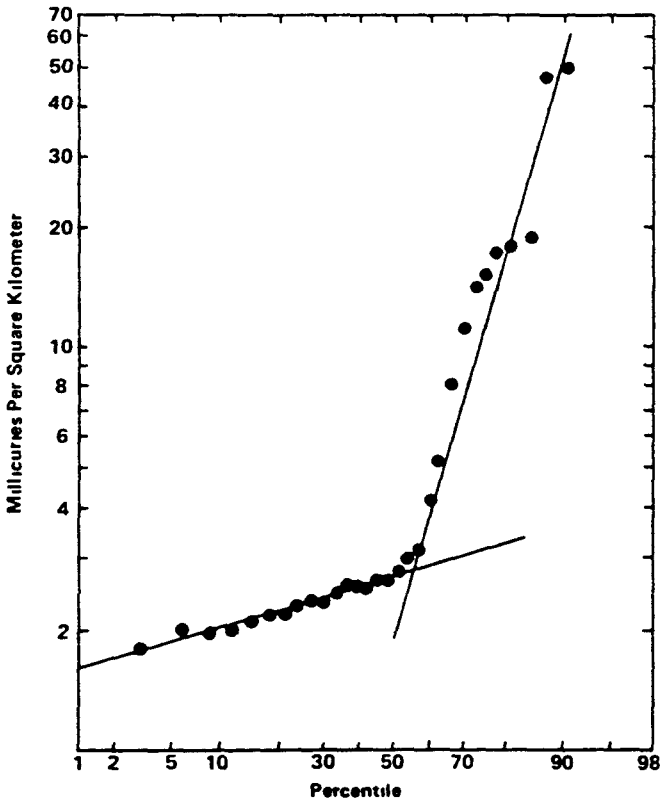


Figure B 1 Compound Distribution Containing Samples Dominated Either by Background Plutonium or By the Rocky Flats anomaly

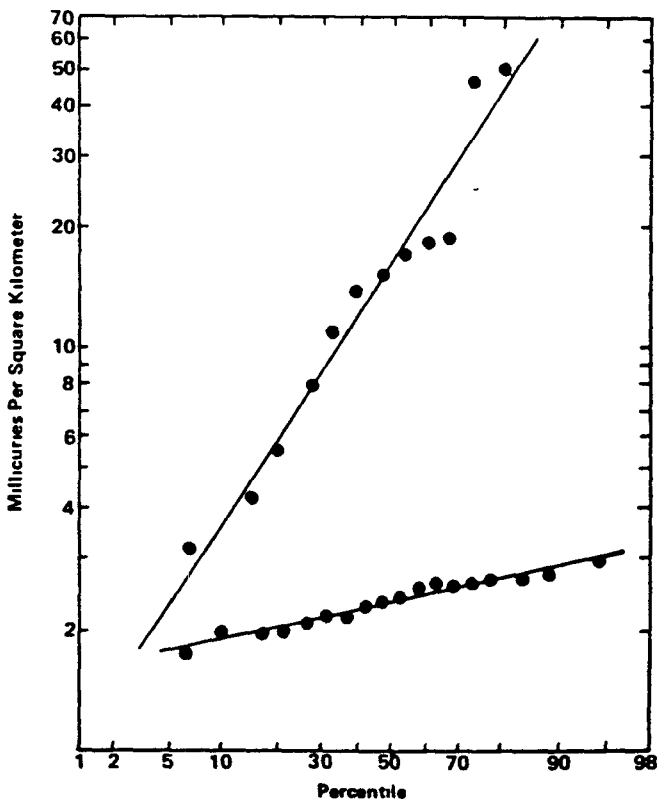


Figure B 2 Background Distribution of Plutonium (lower plot) Separated from the Rocky Flats anomaly

Table B 2 Order and Percentiles for Subgroups

Lower content			Higher content		
Sample	mCi/km²	Percentile	Sample	mCi/km	Percentile
33	1 8	5 3	2	3 1	6 7
18	2 0	10 5	3	4 2	13 3
27	2 0	15 8	11	5 4	20 0
29	2 0	21 0	19	8 0	26 7
31	2 1	26 3	4	11	33 3
10	2 2	31 6	17	14	40 0
22	2 2	36 8	5	15	46 7
26	2 3	42 1	14	17	53 3
1	2 4	47 4	15	18	60 0
25	2 4	52 6	16	19	66 7
28	2 5	57 8	12	47	73 4
9	2 6	63 1	13	50	80 0
20	2 6	68 4	7	480	86 8
24	2 6	73 7	8	630	93 3
21	2 7	79 0	6	1950	–
32	2 7	84 2			
30	2 8	89 5			
23	3 0	94 8			
2	3 1	–			

The difference between the average value of 2 4 mCi/km² and the classification value of 3 0 mCi/km² is a joint consequence of both the natural variability of fallout near Denver and the variations inherent to the sampling and analytical procedures Samples indicating plutonium contents of less than 3 0 mCi/km² contain no increment of plutonium from Rocky Flats In statistical terms the certainty is 95% since the 3 0 mCi/km² value corresponds to the 95th percentile of the background value (and to about the 8th percentile of the Rocky Flats anomaly as plotted)

Section 2

PLUTONIUM FALLOUT IN THE DENVER AREA

Donald E Michels

The log-normal analysis described in Appendix B-Section 1 yielded a value for plutonium fallout in Denver Since the

value is large compared to values used by other investigators (see Table B 3) the following discussion seems pertinent Two points are discussed as follows

Table B 3 Plutonium Fallout Near Denver

Source	Method	Value
Michels[a]	log normal plot of 19 HASL samples	2 4 mCi/km^2
Krey & Hardy[b]	single sample from Derby	1 4 mCi/km^2
CCI I[b]	single sample from Loveland	0 19 mCi/km^2 (d)

[a] Appendix B Section 1

[b] Health and Safety Laboratory report HASL 235 August 1 1970

[c] Colorado Committee for Environmental Information report on Dow Rocky Flats Fire January 13 1970

[d] By conversion 1 d/m/g = 4 5 mCi/km^2

1 Other workers have tended to be conservative and have chosen their background values from the lowest of their analytical values which involved samples remote from Rocky Flats This procedure both biases the selection toward low values and ignores whatever natural variation may exist in the overall background distribution Furthermore the analytical variances involved are not well documented hence the values chosen may be low for reasons independent of the natural variations

2 The plutonium background for the world is known independently from direct chemical analysis because of fission yield estimates maintained by workers at Los Alamos Their 1969 estimate of 500,000 curies of plutonium is equivalent to a world-wide average value of 1 1 mCi/km^2 for a uniform distribution Variations from the world-average values are expected because weather patterns and fission events are not distributed uniformly The magnitude of these world-scale variations are documented for fission products, such as strontium-90, which are more commonly analyzed for than plutonium A latitudinal variation in the ^{90}Sr fallout is documented which shows maximum in both hemispheres near latitudes of 40-50° The northern hemisphere maximum for ^{90}Sr is 2 34 times the world-wide average value Since Denver latitude is 39° N we should not be surprised to find the plutonium fallout there to be more than twice the world-wide average, or between 2 2 and 2 5 mCi/km^2

[redacted]

Presentation at
PROBLEMS ANALYSIS MEETING Rocky Flats
TIME 9 00 am May 25 1971

SOIL CONTAMINATION AND ASPHALT PAD

Speaker Dr James R Seed

Dr Seed has a Ph D from University of California in Physical Chemistry (1964) He is familiar to several members of the panel (H E Roser and D E Patterson) with whom he worked during the official investigation of the May 11 1969 fire at Rocky Flats Some of his early work while in Intelligence in the USAF involved separation and analysis of the actinide elements (primarily protactinium uranium and plutonium) from United States and other nations' nuclear test shots Other USAF experience involved surveillance work on (plutonium and uranium) weapons at Manzano Base Albuquerque, New Mexico His speciality field was radiation chemistry His experience and research at Rocky Flats has resulted in expertise in areas of surveillance, product integrity, corrosion problems in general, plutonium ignition, and the chemistry and physics of plutonium that relates to plutonium handling and storage problems He was assigned by the Operating Board of the Rocky Flats plant as chairman of a special committee to investigate, evaluate, and recommend action relative to the plutonium that accumulated in the soil from the outside "Barrel Storage Area "

Dr Seed is director of Product Research and Development at the Rocky Flats Division

SLIDE 1

[redacted]

A general comment was made that many of the problems that would be discussed throughout the two-day meeting would involve environmental concerns

This slide (Slide 1) was a drawing of the theme that had been adopted by a local high school relative to environmental teaching and action programs

It was pointed out that our plant was approximately 20 years old – working in a technical field that for all practical purposes was only about 25 years old Our learning rate was approximately equal to the rate of the growth of knowledge in this "new" field

As we learned we obviously could look on problems we had faced in the past in light of current knowledge and clearly decide we could or should have acted differently had we known what we know now

Problems of plutonium contamination in soil were discussed in general

SLIDE 2

THERE ARE NO STANDARDS FOR PLUTONIUM IN SOIL

SLIDE 3

PRIMARY GUIDELINE 'MAINTAIN AN ACTUAL DOSE AS NEAR ZERO AS POSSIBLE

SLIDE 4

A PROCESS OF JUDICIOUS DECISION MAKING HAS BEEN EMPLOYED IN THE PAST THULE ACCIDENT, PALOMARES ACCIDENT ETC

The various AEC sites which might face similar problems, such as LASL, Mound, LRL NTS, BNW, Pacific Islands as well as the areas where accidents had occurred, Palomares, Spain and Thule, Greenland were alluded to

With the growth of breeder reactors and fuels reprocessing plants, the problems in this area will continue to grow

The problems associated with establishing plutonium soil standards were touched on

1 particle size
2 retention
3 critical organ
4 resuspension

SLIDES 5 AND 6

The accompanying table was divided into two slides (5 and 6)

Data were presented which showed some "standards" that had been adopted by other countries for plutonium in soil (Some liberties were taken in converting units to make cross reference between various papers a little easier)

Data were also presented in some "reasonable" proposed "standards" for the United States This information can be found in a paper presented at the *International Symposium on Radiological Protection of the Public in a Nuclear Mass Disaster*, June 1968 The paper was authored by R L Kathren of Battelle Northwest The data relating to Criterion I of 10 CFR 140 84 (Financial Requirements and Indemnity Agreements) were also discussed This criterion considers surface contamination values above 0 35 $\mu Ci/m^2$ (approximately 77 dpm/cm^2 or 350 mCi/km^2) over 100 m^2 of property as the *minimum* level of transuranic radio nuclides that would comprise a substantial discharge of radioactive material from its intended place of confinement

SLIDES 5 AND 6

MAXIMUM PERMISSIBLE ALPHA CONTAMINATION

Country	$\mu Ci/m^2$	mCi/km^2 [a]	d/m/gram [b]	Remarks
United Kingdom (Dunster)	0 1	100	22 2	Widespread areas contaiminated with plutonium
United Kingdom	0 1	100	22 2	'Inactive areas '
	1 0	1000	222	"Active areas
Czechoslovakia	0 11	110	24 4	Workplaces after decontamination
France	0 1	100	22 2	Equipment and workplaces in "inactive" areas
	1 0	1000	222	Equipment and workplaces in "active" areas
Poland	0 1	100	22 2	Labs restricted to using 100 μ Ci or less
	1 0	1000	222	Labs permitted to use more than 100 μ Ci
South Africa	0 1	100	22 2	Body, personal clothing, inactive areas
	1 0	1000	222	Equipment and workplaces inside controlled areas.
United States ICC	0 02	20	4 44	Interstate Commerce Commission (Dept of Transportation) pertains to interior of vehicles previously used for transportation of materials
U S S R	0 015	15	3 33	Work clothing and surfaces before cleaning
	0 002	2	0 444	Hands and work underclothing before cleaning
	0 006	6	1 33	Work Surfaces after cleaning
United States [c]	0 04	40	8 8	Urban suburban, recreation areas
	0 4	400	88	Rural truck farming, annual food crops, grazing land, milk-shed, etc
	4 0	4000	888	Rural deep root perennials (e g , nuts, certain fruits)
	40 0	40000	8888	Remote or Controlled desert, forest, fenced or limited access areas

[a] Units used in HASL report No 235
[b] Units used by Rocky Flats in reporting soil analysis (in most cases a specific gravity of one (1) was assumed for conversion of units
[c] Recommended at an International Symposium on Radiological Protection of the Public in a Nuclear Mass Disaster (June 1968)

SLIDE 7

HISTORICAL SEQUENCE

- JULY 1958
  DRUM STORAGE AREA ESTABLISHED DURING SUBSEQUENT YEARS DRUMS WERE CONTINUALLY ADDED PRIMARILY PLUTONIUM CONTAMINATED MACHINING OILS
- 1959
  FIRST DRUM LEAKAGE DISCOVERED–RUST INHIBITOR ETHANOLAMINE WAS ADDED TO DRUMS PRIOR TO STORAGE TO MINIMIZE CORROSION
- JANUARY 1964
  FIRST EVIDENCE OF LARGE SCALE DETERIORATION OF DRUMS REPORTED SOIL CONTAMINATION REPORTED AS INCREASING

SLIDE 8

HISTORICAL SEQUENCE (continued)

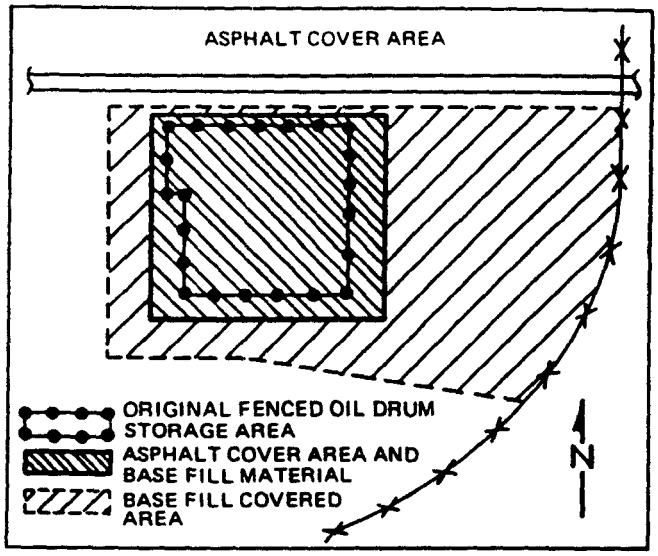
- 1966
  SMALL BUILDING ADDED TO FACILITATE TRANSFER OF CONTAMINATED OIL FROM LEAKING DRUMS TO NEW DRUMS
- JANUARY1967
  LAST DRUMS ADDED TO STORAGE AREA AND REMOVAL TO 774 BEGAN OLDEST DRUMS SHIPPED FIRST
- JUNE 1968
  LAST DRUM SHIPPED TO BUILDING 774 FOR PROCESSING HIGH WINDS SPREAD SOME CONTAMINATION
- JULY 1968
  RADIATION MONITORING AND MAPPING OF AREA COMPLETED LEVELS OF 2×10^5 d/m/gm TO OVER 3×10^7 d/m/gm REPORTED PENETRATION OF FROM 1 INCH TO 8 INCHES REPORTED

SLIDE 9

HISTORICAL SEQUENCE (continued)

- SEPTEMBER 1968
  PRELIMINARY PROPOSAL FOR CONTAINMENT COVER PREPARED BY ROCKY FLATS FACILITIES ENGINEERING
- JULY 1969
  FIRST COAT OF FILL MATERIAL APPLIED
- AUGUST 1969
  FILL WORK COMPLETED PAVING CONTRACT LET
- SEPTEMBER 1969
  OVERLAY MATERIAL SOIL STERILANT AND ASPHALT PRIME COAT COMPLETED
- NOVEMBER 1969
  ASPHALT CONTAINMENT COVER COMPLETED–INCLUDING FOUR SAMPLING WELLS

SLIDE 10



These three slides detailed the significant information relative to the development of the problem and also action taken up to the completion of the asphalt pad

The items on the slides were discussed pointing out that throughout this entire period development work was conducted on a process to treat (dispose reclaim etc) the contaminated oils An initial process was developed in December of 1959 but equipment and funding problems delayed the initial test runs until May of 1960 Further corrosion of equipment problems (hydrolysis and radiolysis of the CCl_4 to HCl) delayed the process until December 1961 This process and project of treating the contaminated oils was included in the project Additional Processing Facilities Contract AT(29-2) 1298' which was an expansion of the plutonium chemical operations Later funding difficulties resulted in deletion of this portion of the project Actual processing of the oil commenced in January of 1967 At this time the field contained about 5 240 drums

In the discussion with the AEC participants of the meeting, the action taken to minimize drum corrosion was covered and the effort made to transfer oil from the older to newer drums was also discussed

SLIDE 10

A drawing of the actual area of the pad and additional base fill was presented and discussed (Slide 10)

SLIDE 11

A photograph of the pad area was presented (Not included in text)

SLIDE 12

A slide which consisted of colored "contours" depicting plutonium in the surface soil around Rocky Flats was presented These data were generated by using soil analysis from

1 Rocky Flats Health Physics Department

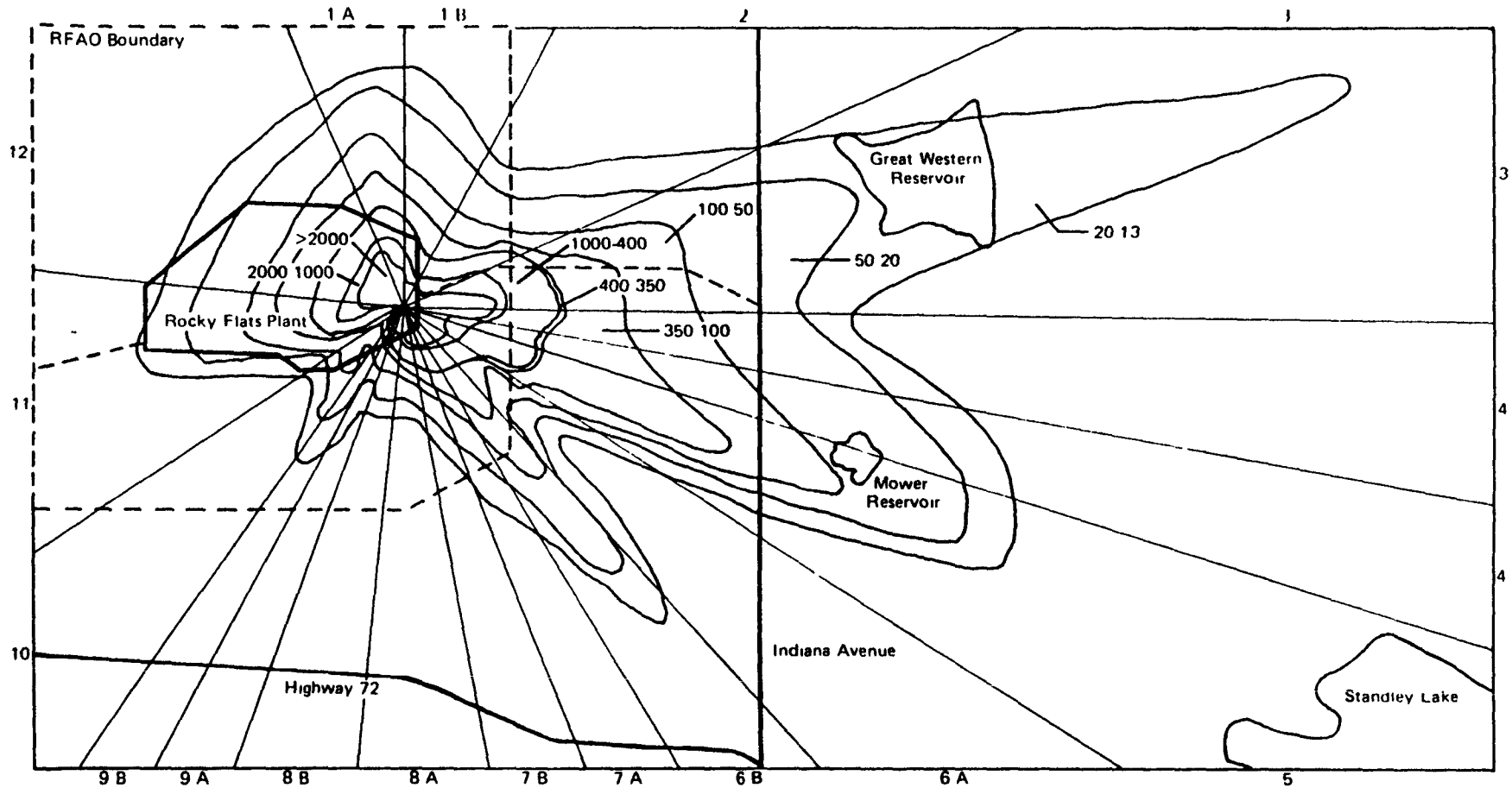
2 The Health and Safety Laboratories (HASL)

3 The Colorado Committee on Environmental Information (CCEI-Martell)

This work had been completed *November 16, 1970*

The model for the "contours" had been constructed from 135 soil sample analyses

1 Ninety-nine samples taken on AEC property and on public and private property by the



Slide 12 An Outline of Rocky Flats Showing the Levels of Plutonium Activity in the Soil in mCi/km^2 Soil Sample Data for the Countours were Evaluated per Sector of the Outline One Inch of the Outline is about 3600 Feet

Rocky Flats Health Physics Department between *August of 1969 and July of 1970*

2 Eighteen samples taken by HASL in *February of 1970*

3 Eighteen samples taken by CCEI in *August of 1969*

SLIDE 13

This slide consisted of another set of 'contours" for plutonium and incorporated into the model Thirty-eight additional soil samples taken during December 1970 (approximately 4 to 6 months after the initial Rocky Flats plutonium soil analysis) This new set of 'contours which was based on a total of 173 soil analyses showed very little change from the previous slide

Additional soil sample analyses showed that the level of plutonium contamination is *not* increasing

The total integrated quantity of plutonium (using simple integral calculus on the equations of the model) was estimated to be 14 3 grams over the 8 35 km^2 (2 063 acres) covered by the orange (1000 → 2000 mCi/km^2) yellow green blue purple brown and gray (13 → 20 mCi/km^2) contour lines (Slide 13)*

There are approximately 6 7 grams over 2 72 km^2 (672 acres) of land inside the boundary below the 2000 mCi/km^2 contour

There are 7 6 grams over 5 63 km^2 (1 393 acres) outside the boundary down to the 13 mCi/km^2 contour

The details and the assumptions that were made in constructing these models can be seen in internal Rocky Flats Reports [redacted]

*In this report Slide 13 is reproduced in black and white tones only

1 Service Report 482 70 2 November 16 1970

2 Service Report 482 71-4 April 26 1971

Copies of the latest report have been transmitted by the Rocky Flats Plant Manager through the local AEC area office to H E Roser Assistant Director, Division of Military Application and J F Burke Assistant Manager for Operations Albuquerque

SLIDE 14

This slide consisted of a table which gave the integrated quantities and land area included in each contour

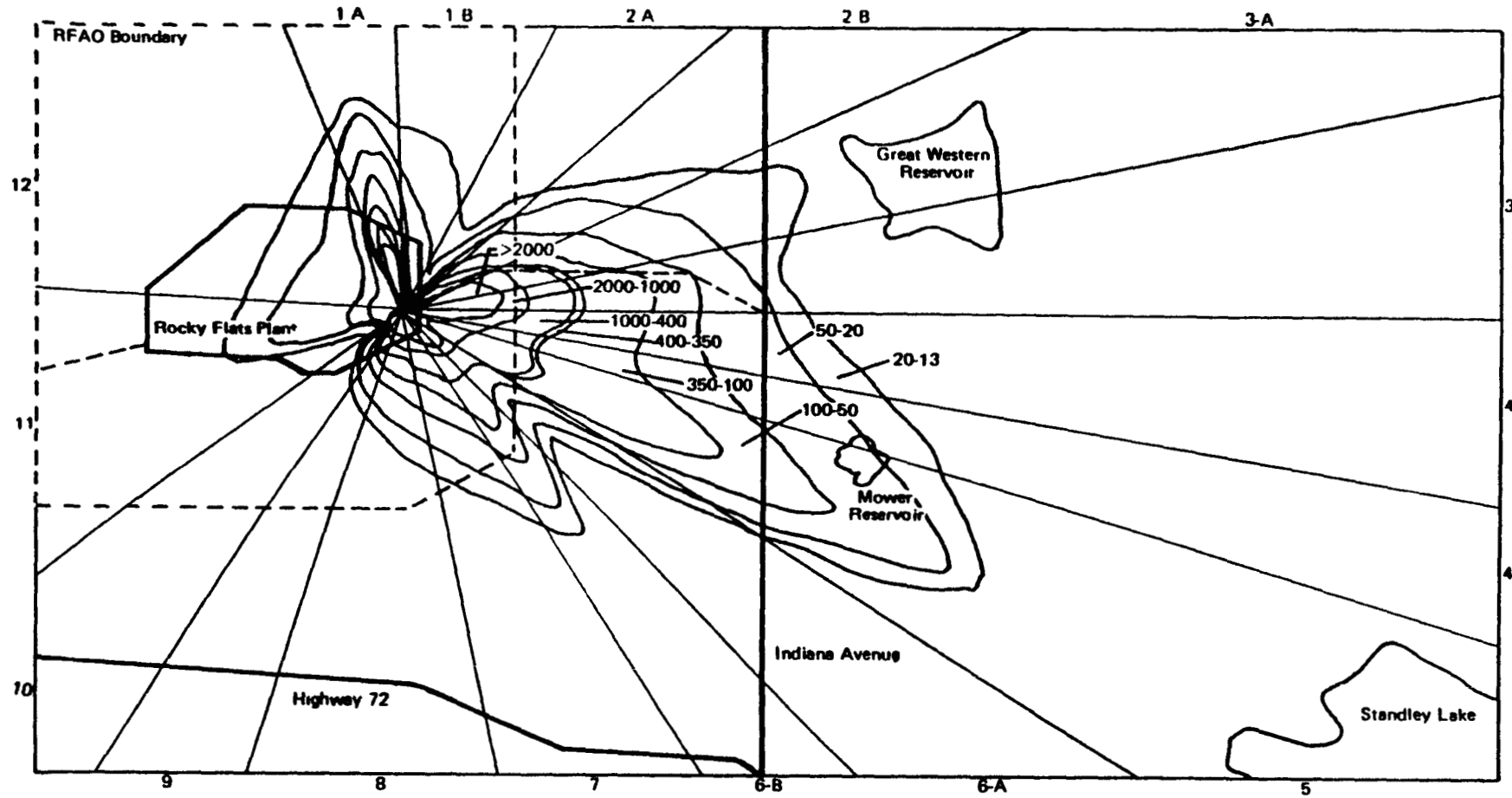
CONTOUR	ACTIVITY IN mCi/km^2		Pu - 239 IN GRAMS	AVERAGE * d/m/g DRY SOIL	AREA IN km^2
ORANGE	2000	1000	0 29 ± 0 02	198	0 02
YELLOW	1000	400	1.25 ± 0 09	131	0 13
GREEN	400	350	0 32 ± 0 03	87	0 05
BLUE	350	100	3 10 ± 0 54	40	1 06
PURPLE	100	50	1 26 ± 0 46	16	1 09
BROWN	50	20	1 04 ± 0 59	7	1.98
GREY	20	13	0 34 ± 0.22	4	1 30
BEYOND GREY	13	1	–	≏.2**	∞
TOTAL			7 6 ± 2 0		

* ASSUMING A DENSITY OF DRY SOIL OF 1 GM/CM³ AND A SOIL SAMPLE DEPTH OF 1 CM

** BACKGROUND PLUTONIUM ACTIVITY

SLIDE 15

This was a photograph of the area of the plant site involved in this discussion and introduced the participants of the meeting to the area they would physically tour later in the morning (Not included in text)



Slide 13 The Recalculated Isodose Contour Lines Showing the Levels of Plutonium Activity in the Soil in mCi/km^2 Soil Sample Data for the Contour Lines were Evaluated per Sector of the Outline One Inch of the Outline is about 3600 Feet

SIMPLIFIED CONVERSION SCALE AND TABLE FOR THE VARIOUS UNITS USED IN THE LITERATURE TO EXPRESS THE LEVELS OF PLUTONIUM CONTAMINATION IN SOIL

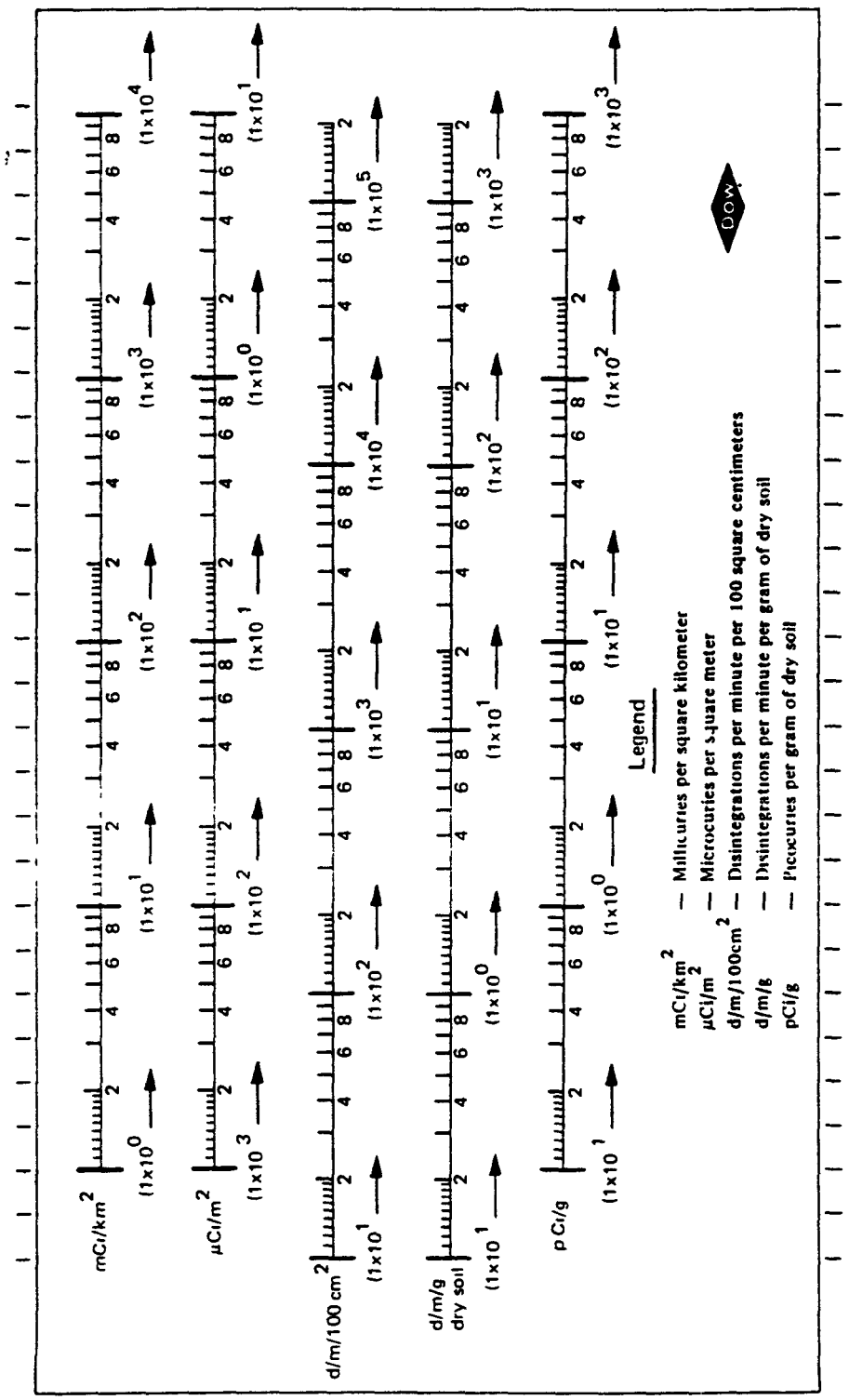


Figure D 1 Simplified Scale for the Various Units used in the Literature to Express the Levels of Plutonium Contamination in Soil

Table D 1 Simplified Conversion Table for the Various Units Used in the Literature to Express the Levels of Plutonium Contamination in Soil

Pu Activity* in mCi/km^2	$\mu Ci/m^2$ *	$d/m/100\ cm^2$ **	d/m/g Dry Soil***	pCi/g†
2000	2 0	$4\ 4X10^4$	$4\ 4X10^2$	$2\ 0X10^2$
1000	1 0	$2\ 2X10^4$	$2\ 2X10^2$	$1\ 0X10^2$
400	$4\ 0X10^1$	$8\ 9X10^3$	$8\ 9X10^1$	$4\ 0X10^1$
350	$3\ 5X10^1$	$7\ 8X10^3$	$7\ 8X10^1$	$3\ 5X10^1$
100	$1\ 0X10^1$	$2\ 2X10^3$	$2\ 2X10^1$	$1\ 0X10^1$
50	$5\ 0X10^2$	$1\ 1X10^3$	$1\ 1X10^1$	5 0
20	$2\ 0X10^{-2}$	$4\ 4X10^2$	4 4	2 0
13	$1\ 3X10^{-2}$	$2\ 9X10^2$	2 9	1 3
1	$1\ 0X10^{-3}$	$2\ 22X10^1$	0 222	$1\ 0X10^{-1}$

*The values are based upon a soil density of 1 g/cm^3 at a depth of 1 centimeter

**The values are based on the relation 1 d/m/g dry soil = $d/m/cm^2$ assuming a soil density of 1 g/cm^3 at a depth of 1 centimeter

***The units in which the results of soil sample analyses are reported.

†The values are based on the relation 1 pCi/g ≈ 2 2 d/m/g dry soil

Note The activity in mCi/km^2 will simply be increased by a factor equal to the depth of the sample, e g 4 centimeters deep will give 4 times the activity assuming a uniform distribution of activity as a function of depth

[redacted]

TRIP REPORT

J R Seed

THE DOW CHEMICAL COMPANY

ROCKY FLATS DIVISION
P O BOX 888
GOLDEN COLORADO 80401

November 9 1970

L M Joshel

TRIP REPORT NEVADA OPERATIONS OFFICE
(PLUTONIUM IN SOIL PROBLEMS)
NOVEMBER 2 AND 3 1970

I Contacts

USAEC

Arthur J Whitman	Radiological Safety Branch
Donald W Hendricks	Radiological Safety Branch
Ross L Kinnaman	Effects Evaluation Office
Paul J Mudra	Operations Division

USPHS – SWRHL

Mel W Carter	Laboratory Director
Jim Mullins	Deputy Chief Analytical Division
Les Dunn	Environmental Survey

REECO (Test Site)

Arden E Bicker	Environmental Services
Derek Engstrom	Chemistry Laboratory
Leonard Sygitowicz	Chemistry Laboratory
Terry Roy	Site Survey

II Topics Covered

A Extent of Test Site Plutonium in Soil Problems "On Site" (REECO + NVO)

B Extent of Plutonium in Soil Problem "Off Site" (NVO + USPHS – SWRHL)

C Soil Analysis and Sampling Problems (REECO + USPHS – SWRHL)

D Soil Stabilization Activities (REECO)

E The Formation of a 'Nevada Applied Ecology Group" – Subcommittee on Plutonium

Chairman	Wright Langham (LASL)
Members	Chet Richmond (LASL)
	W J Bair (BNWL)
	J L Olson (LRL)
	Evan M Romney (UCLA)
	J W Healy (LASL)
	Otto G Raabe (Lovelace)

III REECO Discussions

A Plutonium in soils analysis is being done by normal radiochemical procedures (2 per day at $65 each) and rapid liquid scintillation method (50 per day at $10 each) Mr Sygitowicz will send us a copy of the procedures used for the latter

B REECO at Mercury has a very modern well equipped and adequately staffed radiochemistry laboratory

C REECO uses unique electrodeposition cells, inexpensive and disposable

D REECO is evaluating chemical soil stabilizers as follows

(1) PETROSET (Phillips 66) applied in 1 5 and 1 10 dilutions Their contact at Phillips is Richard Bennett in Bartlesville

(2) DCA-70 (Union Carbide – $1 75/gallon) Applied in 1 40 water dilution Source is Tarrytown Technical Center, Saw Mill River Road at Route 100C, Tarrytown, New York 10591

[redacted]

(3) NORLIG A 11 (American Can Company) 4-cents per pound for powder and 25 cents per gallon for 50% liquid Contact is in Greenwich Connecticut and material is shipped from Green Bay Wisconsin

(4) They have decided to evaluate the Dowell products we are also considering

E REECO expressed interest in an information exchange meeting and soil sample exchange programs

IV PHS - SWRHL Discussions

A Plutonium in soil analysis by standard radiochemical procedures at rate of 2 per day from 1 gram samples for $50 - $75 per sample

B SWRHL has modern well equipped (AEC supplied) and adequately staffed laboratory

C SWRHL plans to collaborate with Claude Sill at NRTS (IDO) in preparation and distribution of "standard" plutonium in soil sample exchange They hope Rocky Flats will be involved

D SWRHL would like to send Rocky Flats cuts from soil samples collected at off-site locations around NTS They also expressed interest in an information exchange meeting on these problems

V NVO Discussions

The people from the area office were very helpful They supplied us with large quantities of reading material on the topic much of which is not generally available Some more detailed reports on "Roller Coaster" and some unpublished follow-up work was very valuable Staff reports on Bikini Atoll relating to rehabitation and plutonium levels in soil were useful One island which will not be rehabitated because of plutonium in soil (83→410 pCi/gm) is Eneman Island *Land that is contaminated with levels similar to that found around Rocky Flats is considered safe for rehabitation* The deepest penetration of plutonium in soil in the Bikini Islands (where the humidity is very high) was found to be 9 inches

A series of assumptions with regard to plutonium in soil on the Islands can be related to Rocky Flats

Assume an activity of 1 pCi/gm Assume *all* activity is on the surface Assume *all* particles one micron in diameter (Actually for effective inhalation particles should be 1 to 10 microns – project Roller Coaster showed 85% of particles to be greater than 10 microns)

Assume *all* particles are *available* for resuspension *initially* Assume 10^{-6} for resuspension factor

Conclusion - Conservatively total lifetime dose due to this plutonium in soil will be less than 70 millirads, a safe level

Project 57 in Nevada showed that the effective half life of high levels of plutonium in soil (based on resuspension data) was as short as 35 days Later studies on lower levels give much longer (unavailable) half lives

Some significant notes taken from the report by the Nevada Applied Ecology Group Steering Committee

1 Any extensive "cleanup" of plutonium contaminated area should not be initiated until extent, health implications and radioecological significance has been evaluated

2 This project should have very high priority "It is *imperative* that the program proceed without interruption " The program referred to is the total evaluation, especially soil sampling coupled with a Fidler instrument (Gamma) type survey

3 Physical and chemical characteristics of plutonium in the environment must be studied, particularly

(a) distribution within soil components,
(b) radioactivity as a function of particle size,
(c) solubility variables

VI Some Additional Reports were of interest

1 "Radiochemical Procedures For The Determination of Plutonium in Environmental Samples," April 1970, Danish AEC Research Establishment, Health Physics Department, Risoe, by Erik Kjaer Markussen

2 "Radiation Characteristics of Plutonium 238," LASL-3696, October 11, 1967, George M Mattack and Charles F Metz

3 "Documentation of Alpha Contamination at the Nevada Test Site," April 1961, unpublished internal

report from REECO to AEC data compiled by B L Brown J L Gardner K H Gunn C H Johnston and R J Scanlon, edited by F W Wilcox

Report presents good summary of 2 year history (1959 and 1960) at four alpha-contaminated sites Data show some alpha particle penetration down to 1⅛ inches and only slight resuspension none to create 'significant inhalation health hazards "

4 NVO-162-28, "Radiological Conditions at Project Roller Coaster 1966, by the staff of Environmental Surveillance Group REECO, January 1967

Conclusions reached were

(a) "Contamination at the sites of Clean Slates 1, 2 and 3 as well as Double Track is well fixed The ground average reading, for the most part is 100 c/m over background "

(b) "It is reasonable to assume that the material has not been resuspended and redistributed On this basis, there does not appear to be any health hazard to the civilian population living in the region at this time '

(c) "It is strongly recommended, however, that the highly contaminated debris exposed by erosion be reburied or otherwise fixed *in situ* ' We now have a copy at Rocky Flats

5 "Alpha Decontamination Proposal - Nevada Test Site and Tonopah Test Range, unpublished copy of a proposal from REECO to NVO-AEC

Proposal discusses study of plutonium contaminated soils at NTS and Tonopah and testing of soil stabilization techniques Suggested materials are DCA-70 and Norlig A The former is a polymer works on the acidic fraction of soils, and application might cost 8 - 12 cents per square yard The latter is a calcium lignosulfate costing about 4 - 6 cents per square yard to apply Total costs are estimated at $335 per acre The data in this proposal were obtained from a U S Bureau of Mines report on work performed on uranium ore tailings at Tuba City, Arizona Actual development work by REECO at NTS is only just beginning with evaluation of several potential stabilizer chemicals

6 BNWL-1221, "Plutonium Inhalation Studies," W J Bair, February 1970 A series of lectures given in Japan in 1969 (Rocky Flats Library has a copy)

7 USBMRI-7288, "Chemical Stabilization of Uranium Tailings at Tuba City, Arizona," Richard Havens and Karl Dean, August 1969 (Rocky Flats Library will order a copy)

8 DA-PAM 525-5, U S Army 1969 Military Operations, "Dust Control - Lessons Learned," no authors listed (Rocky Flats Library will order a copy)

J R Seed
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C T Illsley
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JRS wj

cc
K W Calkins
D Hunt
D T Illsley
W H Lee
F J Miner
J B Owen
K Pocius
C W Piltingsrud
J F Willging